# LOW-RADIOACTIVITY-LEVEL WASTE TREATMENT.

PART II. PILOT PLANT DEMONSTRATION OF THE REMOVAL OF ACTIVITY FROM LOW-LEVEL PROCESS WASTES BY A SCAVENGING-PRECIPITATION ION-EXCHANGE PROCESS

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operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

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ABSTRACT

The results of 16 pilot-plant demonstration runs on the removal of activity from low-activity process wastes by the Scavenging-Precipitation Ion-Exchange process are presented. During an eight month period the runs, ranging from 71 to 146 hr of continuous operation and processing 43,000 to 88,000 gal of ORNL process waste water per run, treated 1.3 million gallons of waste and yielded effluents containing Sr^{90} and Cs^{137} in amounts less than 3% of the maximum permissible concentrations in water (MPC_w) for continuous occupational exposure to these radioisotopes. A standard operating capacity of 2,000 resin-bed volumes was used for the CS-100 carboxylic-phenolic resin column based on its capacity for cesium. At 2,000 bed volumes, the decontamination factors for strontium varied from 2,900 to 12,000 (about 99.99% Sr removal) and for cesium from 246 to 3,444 (99.6 to 99.9% Cs removal). The decontamination factor for strontium decreased to 20 (95% removal) and that for cesium to 6 (82% removal) at 3,118 bed volumes.

The presence of hexametaphosphate in the waste feed had a deleterious effect on the process, resulting in premature fission product breakthrough from the ion-exchange column. Consequently it is recommended that phosphate be excluded from the wastes. Should this approach be undesirable or impractical, sodium carbonate can be used as a process additive to increase resin bed capacity. Several runs indicated that sodium carbonate addition would assure the complete removal of Sr^{90} in wastes containing less than 2 ppm of PO_4^{3-} but would limit Cs^{137} decontamination.

The results of the Pilot Plant and Laboratory work indicate that the process has universal application and will yield an effluent that will not require dilution before ultimate disposal. Process cost studies for a 750,000 gal/day plant indicate a capital cost of $400,000 and a total operating cost of 71.6 cents per 1000 gal. This cost includes chemicals, utilities, labor, maintenance, equipment and building amortization, sludge burial, and evaporation, neutralization, and storage of the resin regenerant in steel tanks. The use of sodium carbonate as a process additive would add 11 cents per 1000 gal to the processing cost.

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1. INTRODUCTION

Low-radioactive-level process wastes originate from sources that include experimental laboratories, cooling water from processing equipment, reactor coolant condensates, decontamination facilities and other sources. These wastes present a difficult disposal problem because of their large volume, making evaporation to solids or storage prohibitively expensive. Since the water must be returned to the environment, treatment of these wastes is necessary where the concentrations of radioisotopes exceed the recommended concentrations for disposal to the environment.[1,2]

At ORNL, about 500,000 gallons a day of low-activity waste is monitored for radioactivity, discharged to the environment if disposal limits are met, or treated to reduce the isotopic concentration to conform with Federal regulations.

The treatment currently given these wastes by the Lime-Soda process does not always afford sufficient decontamination from the hazardous contaminants without depending upon dilution by streams and rivers in the area. The objective of the Scavenging—Precipitation Ion-Exchange Process development program is to reduce the concentration of the radioactive contaminants in the waste to below MPC_w (maximum permissible concentration

in water) levels prior to discharge to the environment, thus providing a process suitable for universal application.

The development of the Scavenging--Precipitation Ion-Exchange Process has progressed through the laboratory and semipilot plant (Part I-ORNL-3322 of this study), and the 600-gal/hr pilot plant is reported in this document. The process removes fission products in two steps: (1) a scavenging precipitation with hydrous ferrous oxide and alkaline earth carbonates followed by solution clarification and filtration, and (2) sorption of the Cs^{137}, Sr^{90} and other isotopes on a carboxylic-phenolic ion-exchange resin.

The pattern of this report is: First, results are reported for 16 demonstration runs in which a total of 1.3 million gallons of ORNL low-activity process waste was treated. Second, the detailed flowsheets and equipment items are described, and their performance discussed. This section is followed by a brief process cost summary. The final section of the report presents conclusions and recommendations based on the pilot plant study. The Appendix contains record tables and computer data.

The authors would like to acknowledge the work done by J. T. Roberts, J. O. Blomeke, J. M. Holmes, and L. J. King of the Chemical Technology Division. The efforts of the Analytical Chemistry Division groups under the direction of E. I. Wyatt, C. L. Burros, and C. E. Lamb are also greatly appreciated.

2. PROCESS DEMONSTRATION

2.1 Comparison of Decontamination Achieved by the Conventional and New Process

At ORNL, process wastes originate from floor drains, fuel-element storage canals, process jackets, coils and condensers, and drains from

the decontamination facility. These wastes are routed through a continu-
ous monitoring station that automatically diverts them either to a
settling basin for untreated discharge from the area, or (if the wastes
contain more than 40 β counts min^{-1} ml^{-1}) to a million-gallon equilization
basin for subsequent fission product removal by the Lime-Soda Process.[3]
(During the period 1961-1962 all wastes were processed through the Lime-
Soda Plant.) Plant effluent is combined with wastes that do not require
treatment and is then discharged to the environment via White Oak Creek,
using the dilution effect of the creek and the Clinch River to reduce the
fission product activity to acceptable MPC_w throw-away limits.

The new process was tested by pumping contaminated waste water from
the suction side of the Lime-Soda Plant into the Low-Level Waste Pilot
Plant (Building 2528). The controlled flow into the pilot plant during
the 16 runs represented 3% of the flow being processed by the large plant;
the total quantity used for pilot plant studies amounted to 1.3 million
gallons, exclusive of that required to obtain system equilibrium.

A comparison of the major contaminants removed by the two processes
over a five-month period (November 1961 to March 1962) indicates a sig-
nificantly better removal of all of the isotopes by the scavenging--
precipitation ion-exchange process. During the period of comparison,
43 million gallons of waste was treated by the Lime-Soda process and 0.6
million was treated in the pilot plant. Table 2.1 presents the results
of the comparison.

Table 2.1. Comparison of Activity Removal During November 1961-March 1962

Process	Isotope Removal (%)			
	Sr^{90}	Co^{60}	Cs^{137}	TRE
Lime-Soda-Clay	62.2	30.8	72.6	72.8
Scavenging--Ion Exchange	>99.9	84.9	98.8	89.7

2.2 Usual Constituents in the Process Waste Water
Entering the Pilot Plant

The feed pumped into the facility was much like ORNL tap water with respect to total hardness, turbidity, and pH. The total hardness of feed solutions ranged from 96 ppm to 138 ppm and averaged 115 ppm (all hardness data is in terms of $CaCO_3$). Turbidity and pH measurements averaged 17 ppm and 7.7, respectively. ORNL produced from 350,000 to 750,000 gal/day of this waste during the pilot-plant runs.

The average radiochemical analysis of composited feed solutions is presented in Table 2.2.

Table 2.2. Average Radiochemical Analysis of
Feed Runs HR-2 through HR-16

Isotope or Equivalent Expressed as β or γ Radiation	Activity	Units
Gross β	36.2	counts $min^{-1} ml^{-1}$
Gross γ	88.5	counts $min^{-1} ml^{-1}$
Sr^{90} β	86.4	dis $min^{-1} ml^{-1}$
Co^{60}	130.2	dis $min^{-1} ml^{-1}$
Ru^{106}	7.7	dis $min^{-1} ml^{-1}$
TRE	9.3	counts $min^{-1} ml^{-1}$
Cs^{137}	54.4	dis $min^{-1} ml^{-1}$
$Zr-Nb^{95}$	< 2.3	counts $min^{-1} ml^{-1}$

2.3 Unusual Constituents in the Process Waste Water
Entering the Pilot Plant

A premature breakthrough of fission products from the resin column during HR-12 prompted a survey of the ionic contaminants being discharged into the ORNL waste system. A concentration of 5 to 6 ppm of Turco 4324 (a commercial decontamination mixture used elsewhere at ORNL and then

dumped into the process waste water), containing some sodium hexameta-phosphate, was detected in plant feed and appeared to have had a deleterious effect on the process. The use of this chemical at the ORNL decontamination facility has been shown[4,5] to inhibit the precipitation of calcium carbonate from water, due to the hexametaphosphate-calcium complex. For this reason, the chemical flowsheet was altered slightly to include the addition of sodium carbonate to induce calcium precipitation; subsequent runs show that the use of sodium carbonate restores the high decontamination of Sr^{90}, but limits the removal of Cs^{137}.

Other contaminants in process wastes included nitric acid, Fab detergent (which also contains hexametaphosphate), ammonium oxalate, acetic acid, hydrochloric acid, Varsol, Versene, Gunk (a degreasing solvent), Amsco, Alamine-336 (a long-chain amine), diethylbenzene, lithium chloride, Prestone, Freon, and lesser quantities of photographic chemicals.

2.4 Constituents in the Water After Treatment by the New Process

In all but two of the demonstration runs (HR-12, HR-14) the average activity of process effluent was less than 3% of current MPC_w (ref 2) values for continuous occupational exposure. The isotopes considered to be the greatest health hazards, Sr^{90} and Cs^{137}, averaged 1.10 and 0.30 percent of MPC_w, respectively. Other principal fission product contaminants, Co^{60} and the total rare earths, averaged 0.17 and 0.35 percent of MPC_w, respectively. Table 2.3 shows the fission product distribution in the feed stream and in the effluent leaving the process.

The effluent averages do not include runs HR-12 and HR-14. During HR-12 the presence of phosphate in the process feed solution resulted in

a hardness breakthrough in a polishing filter and subsequent loading of hardness (principally calcium) on the resin bed. Strontium-90 and Cs^{137}, the two principal radiochemical contaminants which are sorbed on the resin, were present in the plant effluent in concentrations which reached 464 and 8.8% of the MPC_w, respectively.

Table 2.3. Distribution of Fission Product Activities in the
Influent and Effluent Streams of the Process

	Percent of MPC_w[a]					
	Influent			Effluent		
Isotope	High	Low	Average	High	Low	Average
Sr^{90}	7567	2081	3891	2.7	0.45	1.10
Cs^{137}	62	3	12	2.1	0.01	0.30
Co^{60}	149	< 1	12	55	0.06	0.17
Ru^{106}	7	< 1	4	4.41[b]	0.45	1.95
TRE^c	12	3	14	1.1	0.02	0.35

[a]Based on continuous occupational exposure for a 168-hr week.
[b]Runs HR-14, 15, 16 only.
[c]Calculated as Y^{91}; TRE = total rare earths.

During HR-14, the sludge removal rate was too high, causing localized channeling (with consequent breakthrough) in the sludge blanket of the clarifier. This channeling and a polishing-filter breakthrough contributed to a resin-bed breakthrough. Plant effluent reached 153 and 2.24% of the MPC_w for Sr^{90} and Cs^{137}, respectively, at the end of the run (1942 bed volumes).

A Co^{60} spill on the ORNL site during run HR-8 increased the concentration of this isotope in the feed solution to 1650 dis min^{-1} ml^{-1}. This is about 100 times greater than that previously observed. In this case the pilot plant effluent contained 55% of MPC_w.

The MPC_w for Cs^{137} in the plant effluent was not exceeded at any time during the program; however, the maximum concentration observed in feed solutions was only 62% of the MPC_w.

Only three runs (HR-14, HR-15, and HR-16) had sufficient concentrations of Ru^{106} in the feed for accurate analysis. The plant effluent analysis of these three runs indicated an average of 1.95% of MPC_w. Included in this average is the 4.41% which occurred in the abnormal run HR-14.

3. FISSION PRODUCT REMOVAL

3.1 Discussion of the Resin Characteristics and its Affinity for Cesium and Strontium Ions

Sixteen demonstration runs were completed. Volumes of 42,600 to 87,600 gal of process waste water were treated per run, representing 71 to 146 hr of continuous operation. At the 2000-bed-volume level, the plant effluent contained less than 3% of current MPC_w values for continuous occupational exposure,[2] and overall decontamination factors were at least three times higher than those obtained in the laboratory and semipilot plant.[5,6] Decontamination factors for Sr^{90} (Table Al in the Appendix) ranged from 2,900 to 12,000, representing 99.99% removal in up to 2086 bed volumes, and those for Cs^{137} ranged from 246 (99.6%) to 3444 (99.9%) for the same operating period.

During the second run, it was established that the Duolite CS-100 resin bed would function satisfactorily for 500 bed volumes more than the 1,500 predicted from laboratory studies. After 1,500 bed volumes of waste had been treated, overall Sr^{90} and Cs^{137} decontamination factors were 6,143 and 788, respectively. Based on a gamma-radiation scan (Sec

6.4.2) of the resin bed and the absence of a calcium breakthrough, it was decided to extend the on-stream life of the column. After 2,086 bed volumes, decontamination factors from Sr^{90} and Cs^{137} had decreased to 2,047 and 246, respectively, but still represented greater than a 99.7% removal for both isotopes. Two thousand resin-bed volumes were then established as an operational goal for subsequent runs.

The sixth run in the series (HR-6) was made in order to ascertain the resin column breakthrough point, that is, the capacity of the column for cesium and strontium. At 2,000 bed volumes, decontamination factors from Sr^{90} and Cs^{137} agreed favorably with other runs in the series. Cesium was the first to break through at the 50% level after 2,800 bed volumes. At 2,916 bed volumes, Sr^{90} in the effluent was 11.5 dis min^{-1} ml^{-1}, representing 520% of the MPC_w (17% of the Sr^{90} level in feed). Removal factors for Sr^{90} and Cs^{137} from the saturated resin were 3,100 and 1,800, respectively. The removal factor is the peak activity of the isotope during the elution cycle, divided by the activity of the isotope after ten bed volumes of eluant have passed through.

As illustrated in Fig. 3.1, two occasions of accidental Sr^{90} breakthrough were noted (runs HR-12, HR-14). The breakthrough, at the 1% level, occurred after 729 and 1,160 resin bed volumes of waste had been treated. In each of the runs, the total phosphate in feed solutions (2 to 5 ppm) caused excessive total hardness to be delivered to the resin column, thereby decreasing resin capacity. Subsequent runs (HR-15, HR-16) utilized 0.005 $\underline{M}$ Na_2CO_3. The use of this additive has been shown[5] to reduce the detrimental effect of hexametaphosphate concentrations in process wastes up to 3 ppm. Greater than 2,700 bed volumes were obtained in the later runs before the Sr^{90} breakthrough approached the 1% level.
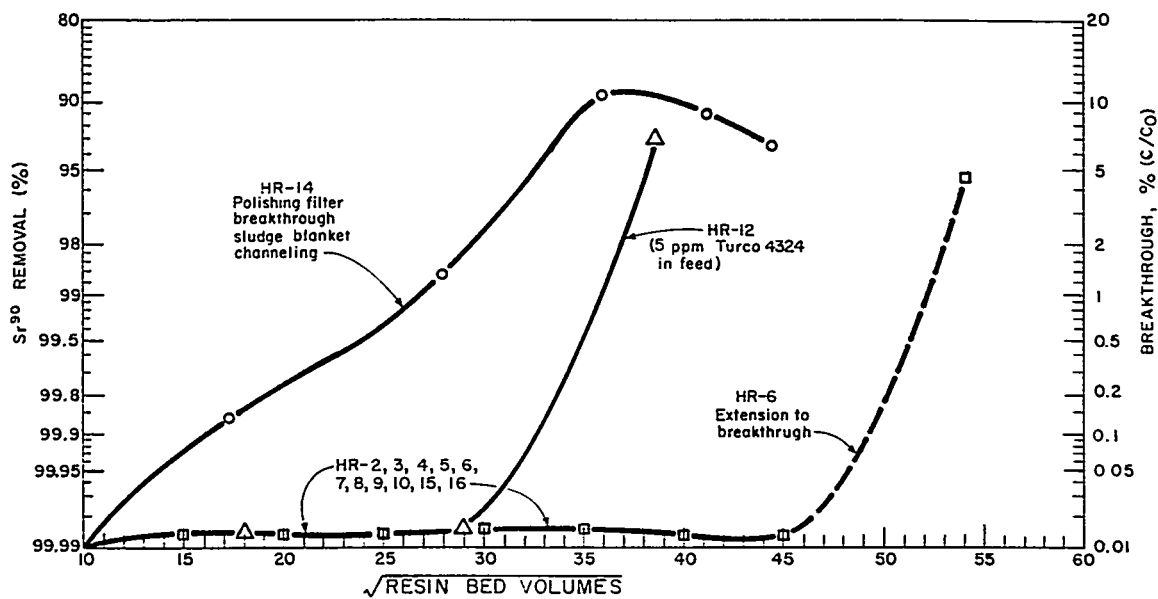
Fig. 3.1. Removal of Sr^{90} from ORNL process waste.

The ability to remove Cs^{137} from the water depended on (1) the pH of the adjusted feed solution, (2) the presence of phosphate in the feed, (3) the total hardness of the solution being delivered to the resin column, and (4) the sodium ion concentration. Referring to Fig. 3.2, runs HR-12, -14, -15, and -16 reached a 1% Cs^{137} breakthrough before the other runs in the series. The withdrawal of sludge during run HR-14 was rapid enough to cause the blanket to channel, thereby causing a resin column breakthrough after 240 bed volumes. Run HR-12 was made under normal flowsheet conditions; however, this was the first run that phosphate was detected in feed streams, causing Cs^{137} to breakthrough at the 1% level after 882 bed volumes. Cesium breakthrough during run HR-16 was caused by an excessive amount of sodium that was added in order to investigate the use of sodium carbonate in the presence of a high pH (11.8). Run HR-15 was made at a somewhat lower pH (11.3), which was undesirable because of the decrease in resin capacity for cesium at lower pH's.

Overall decontamination and removal of the various isotopes may be found in Table A1.

3.2 Degree of Decontamination by the Precipitation and Ion Exchange Steps

Solutions were collected continuously during a run and then analyzed to establish the removal path of each of the fission products in the process. Drip samples representing 0.01% of the total flow were withdrawn from process streams, which included the feed, clarifier effluent, polishing-filter effluent, and the resin-column effluent. The low-activity radiochemical analyses of these samples were evaluated to determine the percentage removal of each of the detectable isotopes across the precipitation-clarification, the filtration, and the ion-exchange steps. The
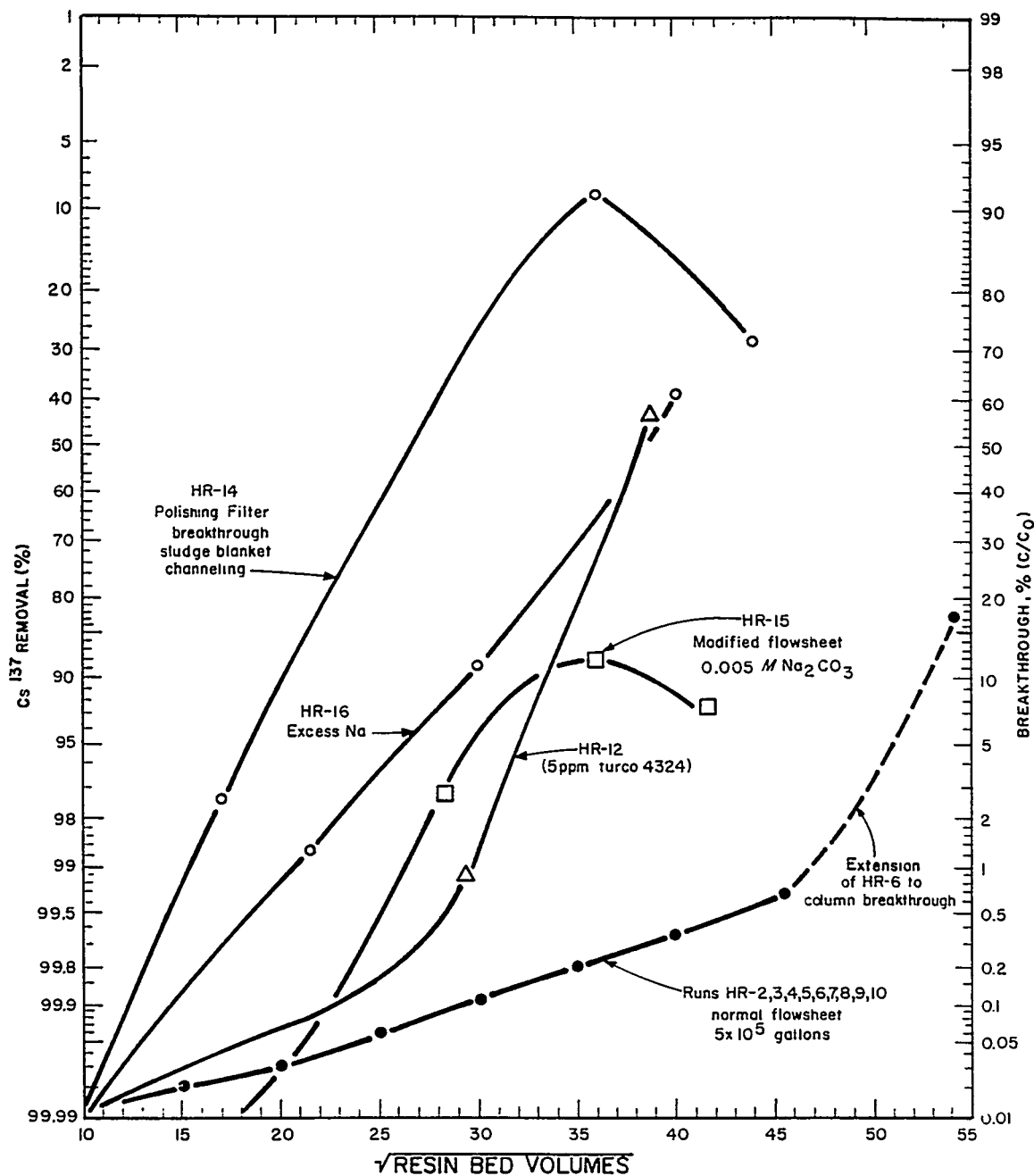
Fig. 3.2. Removal of Cs^{137} from ORNL process waste.

results for each run are tabulated in Table A5 in the Appendix. When no interfering contaminants are in the waste water (Sec 2.3), the clarification and filtration steps remove about 66% of the Sr^{90} and 11% of the Cs^{137}; ion-exchange removes about 99.9% and 99.7% of the remaining Sr^{90} and Cs^{137}, respectively, for 2,000 resin-bed volumes.

4. RADIATION LEVELS IN THE EQUIPMENT AND PIPING

Average radiation data for the process was obtained from Health Physics survey charts of runs HR-3, -4, and -5. These combined runs represent 269 hr of plant operation. Radiation readings were taken at the point of contact between the equipment and the radiation-detection instruments unless otherwise specified.

Maximum readings for the process feed line averaged 0.23 mr/hr. The overall maximum was 0.40 mr/hr; the overall average was 0.19 mr/hr.

The average maximum reading at the flocculator was 7.0 mr/hr, the overall average being 3.9 mr/hr. These readings were taken at the top of the flocculator at about 0.5 to 1 in. from the surface of the liquid. Solvents and detergents in the waste feed, along with the sodium hydroxide introduced to the system in the flash mixer, formed foam in the flocculator after overflowing from the flash mixer. Strontium, having a strong affinity for the foam surface, concentrated in the foam, thereby producing a higher-than-normal radiation field. The overall maximum reading for the flocculator was 13.0 mr/hr.

A permanent beta-gamma detector continuously monitored the intake line of the clarifier. The average reading was 3,900 counts/min and the maximum was 7,700 counts/min. Three readings were taken on the side of

the clarifier at 1, 3, and 4.5 ft from the bottom. The maximum and average readings of the three positions from top to bottom were 0.9 and 0.2, 1.8 and 1.1, and 5.0 and 2.4 mr/hr, respectively. A survey was also made at the top of the clarifier at about a half to an inch from the surface. The maximum and average readings were 0.5 and 0.25 mr/hr.

The polishing filters had maximum and average readings of 2.0 and 1.2 mr/hr, respectively, during the above-stated runs. However, contamination buildup in later runs produced maximum readings of about 10 mr/hr. When the top of one of the anthracite filters was removed so that the filter could be replaced, 40 mr/hr was recorded.

The sludge concentrates radiochemical contamination, and when the sludge filter was in operation, a maximum of 0.5 mr/hr was reported. However, when the filter was open, the filter bag read as high as 12.0 mr/hr, the average being 5.7 mr/hr. The sludge is stored in 55-gal drums for burial. During a run, the drums, which hold about eight full filter bags, were placed in the building storage area. In the two or three days that the drums were in the area, their surface readings increased to 10 or 15 mr/hr. The drums were next placed outside the building in a radiation zone. After remaining in this area for a few weeks, readings as high as 400 mr/hr were reported (this area had a 20 mr/hr background).

Samples of the resin column eluate were monitored throughout the operation. The maximum reading of the first five bed volumes of all runs, HR-2 through HR-16, was 8 mr/hr; this occurred during run HR-8. The average maximum reading of these runs was 3.5 mr/hr. The second five bed volumes, which is stored for use in the split-elution cycle, were not above background. The scanning of the resin column to establish the column breakthrough is discussed in Sec 6.4.2.

The high Co^{60} content of the waste feed during HR-8 raised the radiation level of all pilot-plant equipment to unusually high ones. The background in the area of the feed line was 4.0 mr/hr. There was a reading of 20 mr/hr at 1 ft above the liquid surface in the clarifier, and readings of 6.5 and 9.0 mr/hr were reported for the top and bottom positions, respectively, on the side of the clarifier. The surge tank reading was 20 mr/hr, and the background from the polishing filters was about 6. The resin column reached 20 mr/hr at the 6-in. level. The background of the area near the resin column was 6.0 mr/hr, the highest for this area during any run.

5. THE FLOWSHEET: BASIC STEPS AND UNDERLYING CHEMICAL PRINCIPLES

The flowsheet for the process is shown in Fig. 5.1 and 5.2. Ion exchange with phenolic cation-exchange resins was chosen for evaluation because the phenolic group is highly selective for cesium and strontium, though the useful capacity is large only at pH's high enough to cause a significant fraction of these phenolic groups to be ionized.[6] Since process-water wastes are generally neutral it is necessary to make the waste 0.01 $\underline{M}$ in NaOH to raise the pH to about 12, which is near optimum for cesium and strontium removal from dilute solutions by phenolic resins. When typical ORNL low-activity waste water is made alkaline, most of the dissolved calcium and magnesium precipitate carrying down with them the dirt, algae, and other suspended solids, and a substantial fraction of the radioactivity to such an extent that filtration removes about 1 to 10% of the cesium, about 30 to 70% of the strontium, about 70 to 90% of the ruthenium and cobalt, and more than 60% of the rare earths. In the ion

PROCESS WATER WASTE
1500-2000 RESIN BED VOLUMES
PER 3.5-7 DAY CYCLE

0.5 vol DISPLACEMENT RECYCLE

2

0.5 vol RINSE RECYCLE

3

20 vol BACKWASH RECYCLE

4

1

10.0 vol ACID ELUATE WASTE

NaOH TO MAKE
WASTE 0.01 M

COPPERAS TO MAKE
WASTE 5 ppm IN IRON

POLISHING
FILTER

PHENOLIC
CARBOXYLIC
CATION
EXCHANGE
RESIN
(1 vol)

5.0
VOL

TO WASTE
EVAPORATOR

FLASH MIXER

FLOCCULATOR
~30 min HOLDUP

CLARIFIER
2-3 hr HOLDUP

ELUANT
10 vol 0.5 M HNO$_3$

CONC. TECH. HNO$_3$

WATER

~3 vol SLUDGE

DECONTAMINATED
EFFLUENT

WATER RINSE
1 vol

FILTER

BACKWASH
20 vol 0.1 M NaOH

~0.3 vol WET CAKE

DISCHARGE

~0.1 vol DRY CAKE

STORAGE

Fig. 5.1. Process water decontamination with carboxylic-phenolic ion-exchange resin.
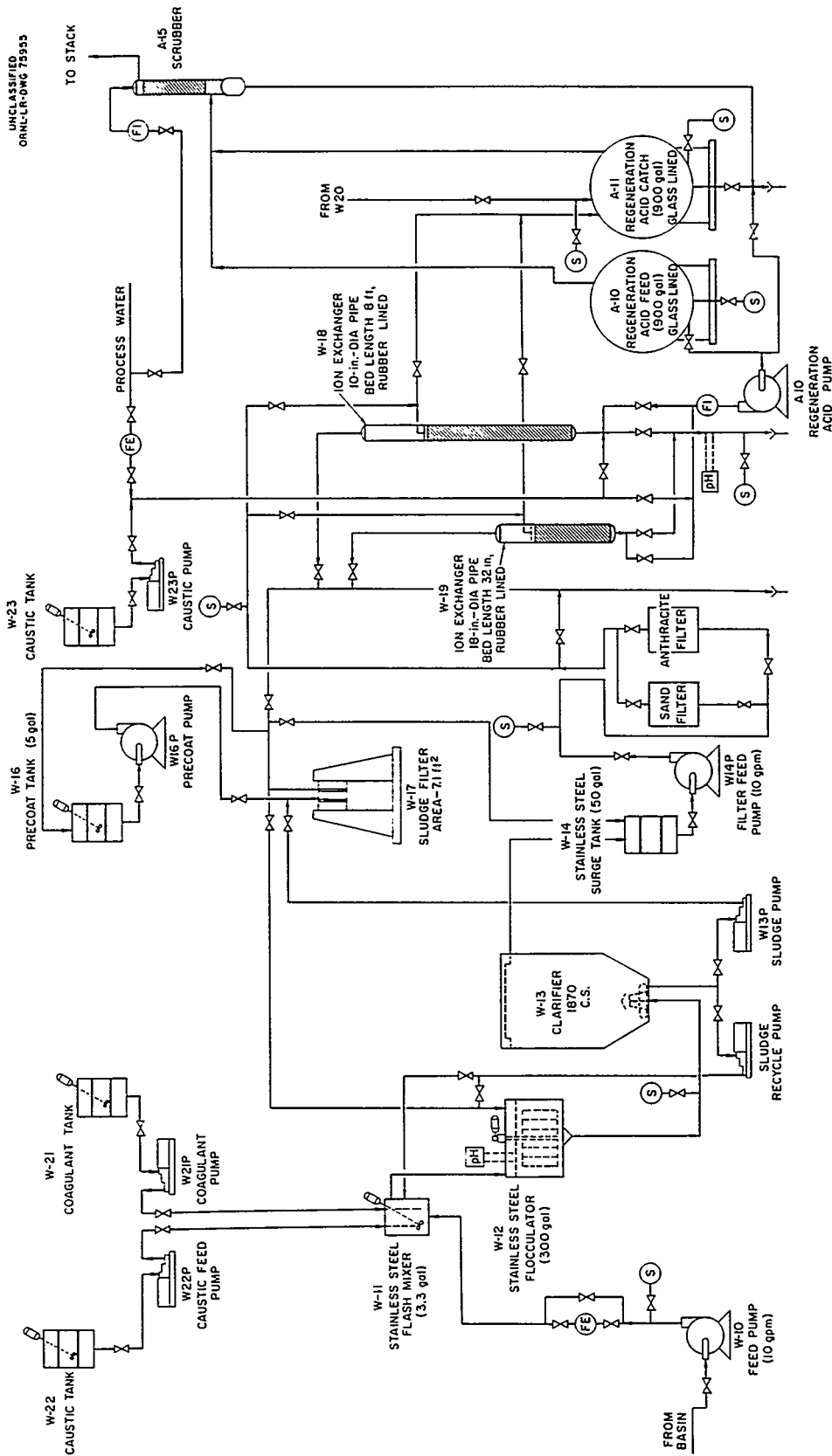
Fig. 5.2. Flowsheet for low-level waste-treatment pilot plant.

exchange step that follows the scavenging precipitation, clarification, and polishing filtration steps, 1,500 to 2,000 volumes of waste are treated with one volume of phenolic resin on a three-day exhaustion cycle with decontamination factors of about 10 for ruthenium and cobalt, and 100 for rare earths, 100 to 1,000 for cesium, and greater than 1,000 for strontium. Fortunately, the radioactive ions for which ion-exchange is not very effective (e.g., ruthenium, rare earths, and cobalt) are precipitated, and those that are only partly scavenged (cesium and strontium) are effectively removed by ion-exchange. Thus, it is not difficult to reduce the major biological hazards to less than 10% of the present MPC_w for environmental discharge by means of the integrated precipitation-clarification ion-exchange steps.

In the head-end or precipitation and filtration steps, it is necessary to reduce the soluble calcium and magnesium concentrations to a minimum because these ions compete with strontium and cesium for exchange sites and can cause premature saturation and breakthrough of the ion-exchange resin. Recirculating the sludge aids in the reduction of soluble-hardness ions (often called "breaking-hardness supersaturation") by contacting the freshly treated waste with preformed precipitates.

The coalescence of small flocs into larger ones reduces the surface area available for the seeding effect; therefore, the hardness is broken best in the flash-mixing stage before flocculation occurs. The kinetics of the precipitation reactions are improved by increasing the temperature of the water from the normal 11 to $18^{\circ}C$ to the optimum of about $25^{\circ}C$, but the added costs of heating the waste compared with savings in size of equipment are probably uneconomical.

The presence of hexametaphosphate in the waste also interferes with the precipitation of calcium carbonate by reducing the surface area of the carbonate precipitate available for seeding and by forming soluble complexes. However, this detrimental effect can be overcome by adding sodium carbonate in the headend treatment, during periods of increased consumption of detergents and decontaminating solutions that contain hexametaphosphate.

Finally, the solids produced must be removed prior to ion-exchange since the resin bed is not an efficient filter. The bulk of these solids are removed nicely with a sludge blanket or suspended-solids contact clarifier. The addition of 5 to 10 ppm of ferrous iron is desirable for floc formation, particularly during periods of low hardness in the feed and when algae concentrations are high, because the iron adds the necessary bulk for smooth sludge-blanket operation.

The solids collected in the clarifier are withdrawn either continuously or periodically and dewatered by pressure filtration in preparation for disposal by burial. The clarified effluent is passed through a polishing filter, either sand or anthracite beds, to remove the last traces of particulate matter and associated radioactivity.

After exhaustion of the resin capacity in the loading cycle, the activity is eluted from the resin with ten resin-bed volumes of 0.5 $\underline{M}$ HNO_3.[6] The resin is then washed with water and regenerated to the Na^+ form by a 20-resin-bed-volume backwash with 0.1 $\underline{M}$ NaOH. The acid eluant requirement is halved by employing split elution, in which case, the second five volumes of acid (very low in radioactivity) are held for use as the first five volumes in the subsequent elution cycle. Recent work shows that the first five volumes in each cycle can be volume-reduced by

evaporation of either the acid or neutralized form. Volume reductions of at least 30 to 1 have been demonstrated in this step, producing less than 0.2 of a bed volume of concentrated waste for disposal. This concentrated waste, plus the 0.1 to 0.3 of a bed volume of solids from the head-end precipitation step, shows an overall process volume reduction from original waste water of greater than 3000.

6. DESCRIPTION OF THE OPERATION OF THE PILOT PLANT

6.1 Introduction

The process consists of three basic operations: precipitation, filtration, and ion exchange. The first two of these, precipitation and filtration, utilize conventional water-treatment equipment, namely, a flash mixer, coagulator, clarifier, and filters. The ion-exchange equipment requires (for the wetted surfaces) a material of construction resistant to sodium hydroxide and nitric acid; equipment lined with polyvinyl chloride (PVC) was chosen to permit the future demonstration of the hydrochloric acid flowsheet. All piping in the ion-exchange system is unplasticized PVC schedule-80 pipe joined with screwed fittings; PVC ball valves were highly dependable in this service. Carbon-steel pipe and valves are used throughout the precipitation and filtration systems, except for one stainless-steel line required for the occasional acid backwashing of the sand and anthracite polishing filters.

The mechanical performance of the equipment under continuous operating conditions was highly satisfactory, requiring very little downtime for maintenance and repairs. Low levels of radiation associated with this process did not require the remote operation of any equipment.

6.2 Precipitation Step

The partial precipitation of the carbonates is accomplished in three steps (some of the strontium carbonate, for example, is not precipitated): mixing, flocculation, and clarification by the sludge blanket method. In modern practice all three steps would be performed very efficiently with a minimum of piping in one large piece of equipment, called a precipitator. However, to facilitate the gathering of accurate chemical and physical data, each step was performed in a separate vessel equipped with sample ports and, where necessary, read-out instruments for continuous recording of data. The vessels have ample holdup capacity allowing sufficient time for the various reactions to reach equilibrium, and all were open to the atmosphere, permitting visual inspection.

6.2.1 Mixing

A 1.5-hp centrifugal pump transfers contaminated waste water at 10 gpm from the equalization basin into the flash mixer (W-11). This mixer is a 5-gal Alsop mixer modified with 1.5-in.-IPS bottom inlet, and side-outlet nozzles, to give a 3-gal holdup capacity. The object of the flash mixer is to mix the waste water with sufficient volumes of NaOH and $FeSO_4 \cdot 7H_2O$ (Copperas) solutions to result in a final solution that is 0.01 $\underline{M}$ in OH^- and 5 ppm in Fe^{2+}. The reaction of the chemicals is quite fast, and the more rapidly they are mixed with the low-activity waste, the more efficient is the use of a coagulant, in this case the Copperas. Thorough and rapid mixing, essential for uniform precipitation throughout the water, is achieved with a variable-speed agitator having a 0- to 1,725-rpm range. During the short retention time (18 sec), only small particles of floc,

consisting of light precipitates of calcium, magnesium, and iron compounds are formed in this initial precipitation step prior to overflowing into the flocculator.

6.2.2 Flocculation

While the chemical reactions that initiate precipitation occur almost instantaneously, the coagulation of the precipitates into large particles is relatively slow, without agitation. Coagulation is accomplished in the flocculator (W-12), where additional precipitation and the formation of larger flocs are obtained by properly controlled agitation.

The flocculator is a modified Dorr thickener, 4 ft in diameter and 3 ft deep, having a holdup capacity of 280 gal. Six vertical wooden blades supported between upper and lower horizontal beams are rotated by an adjustable-speed drive. The blades are turned at 45° angles to the supporting beams and cause a gentle rolling action in the water as the agitator rotates. Two vertical wooden baffles installed 180° apart on the inside of the vessel walls prevent bulk rotation of the water. The pH control of the water is accomplished with two submerged electrodes - an I&N 1199-37 measuring electrode and an I&N 1199-35 reference electrode; read-out is on a Foxboro pH Dynalog recorder with high and low alarm contacts to warn operators of sudden changes in the pH.

During the 30-min hold-up period, the correct agitation rate is highly essential for the coagulation of small particles into large ones; gentle agitation is conducive to good floc formation, while violent agitation prevents it. An optimum agitator speed range between 1 and 1.5 rpm (equivalent to a peripheral speed of 12 to 18 fpm for the outer paddle blades) gave the desired rolling action, which greatly aided the

coagulation of the heavier floc so necessary to the sludge-blanket method
of water clarification.

6.2.3 Clarification

The floc flows out of the bottom of the flocculator and into the
bottom of the clarifier (W-13), in which the particles are separated from
the water by the sludge-blanket method. The clarifier is a modified
design of a vertical Spaulding precipitator from which the mixing zone
and the agitators have been eliminated. The floc settles in this large,
open-top, 1870-gal vessel during a 3-hr retention period. The lower half
of the clarifier has the shape of an inverted, truncated cone, and the
upper half is cylindrical (Fig. 6.1).

The floc entering the bottom of the clarifier strikes an inverted
cone-shaped baffle, deflecting the water outward into a section of the
vessel that has a cross-sectional area of 22.3 ft^2, thus reducing the
velocity from 1.1 fps to 2.2 fph. The heavier particles begin settling
immediately, and the lighter ones begin flowing upward at a gradually
decreasing rate through a suspended bed of particles, commonly called a
"sludge blanket."

This operating principle of clarifiers is well known. It involves
the use of a sludge blanket, held in suspension by the velocity of the
water passing upflow through the clarifier. The sludge blanket not only
functions as a filter by stopping and coalescing with other particles
passing up through it, but it also furnishes additional nuclei to aid in
the precipitation of the insoluble carbonates (by crystal growth), and
the scavenging of the radioactive contaminants (through adsorption).
As particles throughout the sludge blanket agglomerate into larger masses,
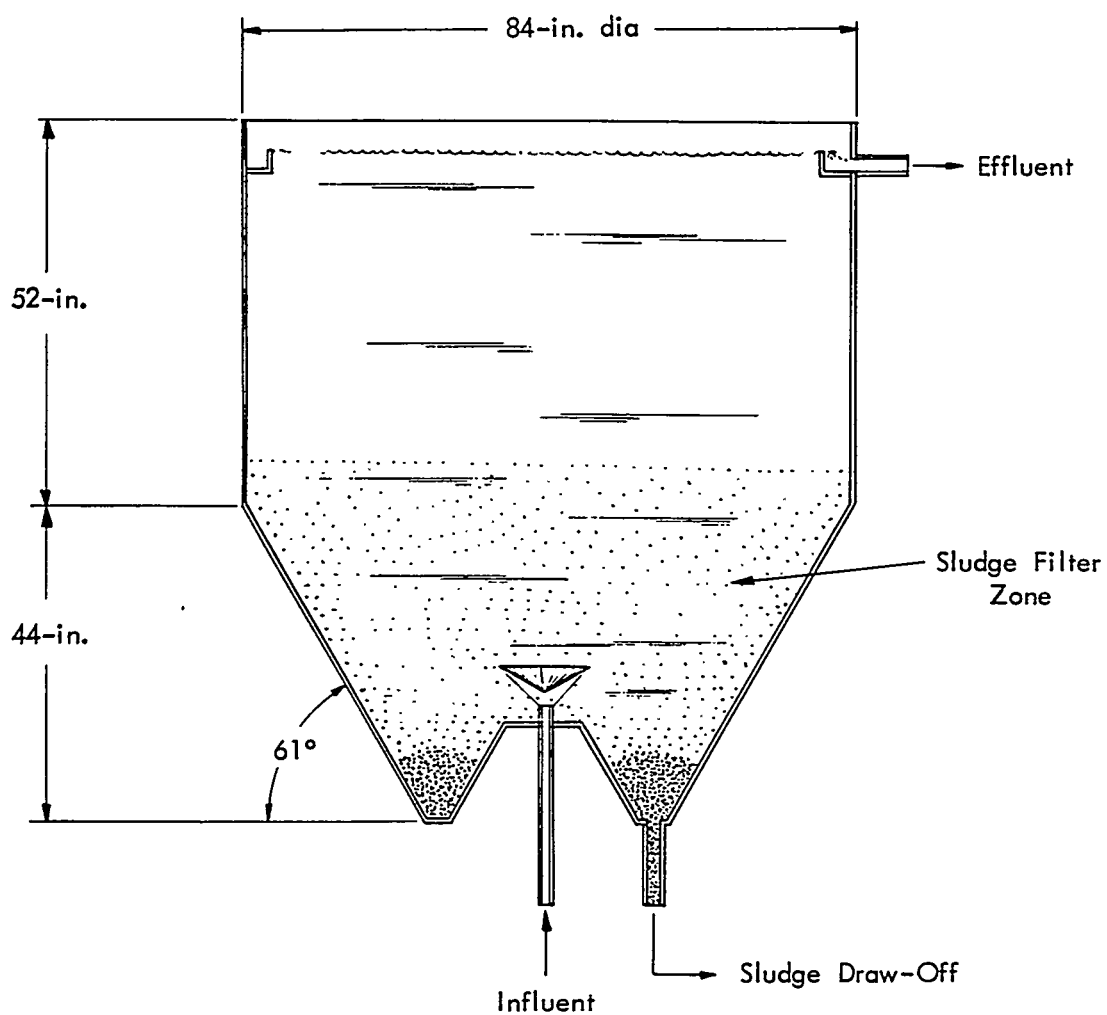
Fig. 6.1. Schematic cross-sectional view of clarifier, W-13.

they settle lower in the water until finally they form a slurry on the
bottom, at which point they are withdrawn from the clarifier and compressed
into a cake by pressure filtration. The volume reduction is about 2,500.

The clear upflowing effluent overflows into a collection trough and
then cascades into a 55-gal-drum surge tank (W-14), from whence it is
pumped through the polishing filters prior to ion-exchange treatment.
Optimum reduction of the effluent hardness is quite important because the
on-stream life of the polishing filters is greatly increased by decreasing
the hardness of the effluent. Supersaturation of the effluent with calcium
and other metal carbonates resulted in postprecipitation and the formation
of a whitish scale on the piping and equipment. The addition of 25 ppm of
slaked lime to the flash mixer (W-11) reduced the hardness from 70 ppm to
56 ppm, but the resultant floc was not as dense, making control of the
sludge blanket level difficult. Recycling of sludge at various rates
from the bottom of the clarifier to the flocculator to provide additional
nuclei did not decrease the hardness but did result in a denser sludge
blanket with better filtering characteristics. Although laboratory work
indicated that increasing the temperature of the feed water from $18^{\circ}C$ to
$24^{\circ}C$ would decrease the hardness, this was unsuccessful in the pilot plant.
The addition of 0.005 $\underline{M}$ Na_2CO_3 to the flash mixer was effective, reducing
the effluent hardness from 60 ppm to 20 ppm, thereby increasing the on-
stream life of the polishing filters by 52%.

The occasional channeling of the influent stream up through the sludge
blanket permitted a portion of the carbonate-supersaturated water to bypass
the sludge blanket, resulting in a sharp increase in the hardness of the
effluent. Channeling usually coincided with high rates of sludge withdrawal

from the sludge-concentration section in the bottom of the clarifier. The small holes in the sludge-withdrawal header were easily plugged by the small, white, carbonate precipitate, causing erratic sludge-withdrawal rates and varying sludge densities. The header was replaced by four nozzles (1-in. pipe), equally spaced around the bottom of the sludge-concentration section and each one valved to permit the controlled drawoff of sludge from any quarter section of the sludge-concentration section. The original cone-shaped baffle was positioned to deflect the incoming influent downward toward the sludge-concentration section; however, the resulting turbulence would not permit the sludge to thicken sufficiently for good filtration. Also, the turbulence in the cone broke the large, rapid-settling floc into smaller, slow-settling particles. However, an inverted cone accurately positioned over the inlet nozzle eliminated the turbulence in both sections, and the original method for baffling was discarded.

Figure 6.2 shows the precipitation, filtration, and mixing equipment used for the demonstration.

6.3 Sludge Filtration Step: Discussion

Since a large portion of the radioactive contaminants in the feed stream is scavenged in the sludge, volume reduction is essential in packaging and disposing of the contaminated sludge. Filtration is the most practical way of concentrating the sludge. However, the presence of algae, organic liquids, and detergents in the low-activity feed that entered the pilot plant resulted in a dewatered sludge that was slimy and difficult to filter.
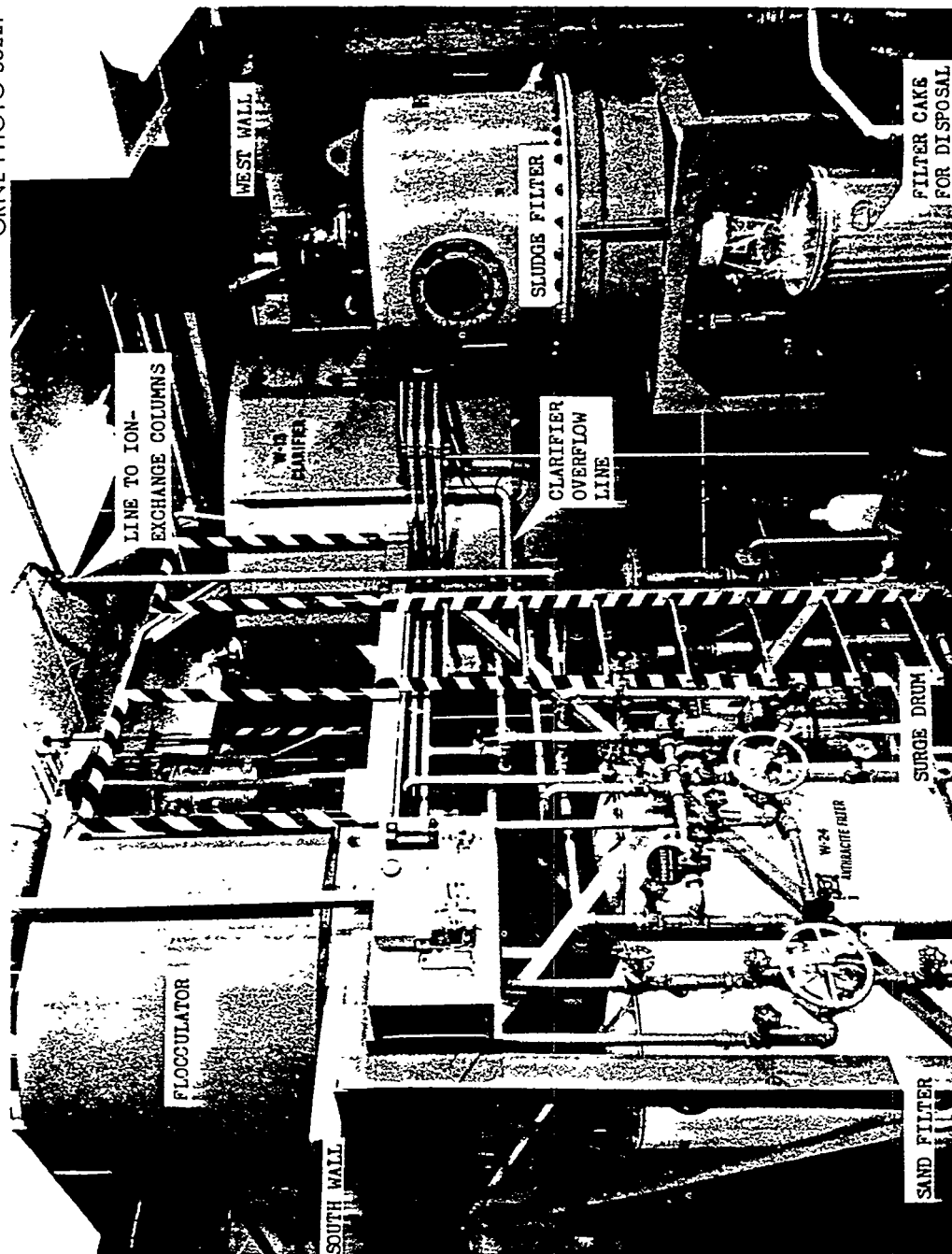
Fig. 6.2. Low level waste treatment pilot plant — pretreatment equipment.

Two methods of filtering this sludge were demonstrated in the pilot plant: (1) vacuum filtration with a rotary drum vacuum filter, and (2) pressure filtration with a plate-and-frame pressure filter. Pressure filtration had the following mechanical and operational advantages over vacuum filtration:

1. Better volume reduction and a more compact cake.

2. Required no auxiliary equipment such as vacuum pump, vacuum condensate receiver, condensate draw-off pump, and a variable-speed drive.

3. No off-gas problem of handling oil and water vapors from vacuum pump.

4. No deleterious effects from erratic sludge composition and feed rate.

5. Shorter precoat time and no excess waste of precoat material.

6. Had no continuously moving parts - hence, less maintenance.

7. Required less attention.

In lieu of the enclosed-bag method (Sec 6.3.2) developed in the pilot plant, the only disadvantage would be in the additional containment required around the pressure filter.

6.3.1 Vacuum Filtration

An Oliver rotary-drum vacuum filter was really not suitable for this particular type of sludge, though it was used in cold runs and in the first four hot runs. The filter, a laboratory type having 3 ft^2 of filtering area enclosed within a pressure hood, was not equipped with a micrometer-drive mechanism for advancing the knife into the precoat, as is done

normally with this type of filter. The rigidly positioned knife permitted
a 1/4-in. layer of precoat material over the No. 120 unbleached cotton
duck that covered the drum. Water washing of the cake is not required;
therefore, all division blocks were removed to permit a longer drying
time per drum revolution. Two grades of diatomaceous earth were used as
precoat and filter-aid materials, Standard Super-Cel and Celite 512.

During shakedown and cold-testing, the filter performed well on ORNL
tap water, which was used as a stand-in for process waste water. However,
this satisfactory performance could not be achieved during the first four
hot runs because of the different characteristics of the sludges formed
by tap water and by waste water. The pores of the precoat were quickly
clogged by the slimes, forming an impervious layer through which the
available pressure drop across the drum (25 to 28 in. Hg) was insufficient
to force water at an adequate rate. Operation was not improved by varying
the precoat thickness, the feed-to-filter-aid ratio, and the drum-rotation
speed.

Suitable operation of this filter could have been obtained by pres-
surizing the pressure hood to increase the pressure drop (driving force)
across the precoated drum. (This would have required secondary containment
around the unit to meet Radiation Safety and Control requirements.) The
addition of the micrometer-drive mechanism would also have made this filter
operable. However, the thorough draining and cleaning of the slurry tank
prior to the necessary re-precoating operation for each filtering cycle,
would have resulted in a longer off-stream time, more attention from the
operators, and a higher consumption of precoat and filter-aid materials
than is economically desirable for this type of plant. For these reasons,

a moving-belt vacuum filter would be more desirable than the rotary-drum filter in this process. A photograph of the dense and undesirable sludge being removed from the process by vacuum filtration is shown in Fig. 6.3.

6.3.2 Pressure Filtration

Pressure filtration was a very effective and convenient method of concentrating the contaminated sludge. This method was demonstrated with a single-frame Eimco-Burwell plate-and-frame pressure filter, having 7.5 ft^2 of filtering area.

The messy filter-cake-removal problem, normally associated with this type of filter, was eliminated by designing a canvas bag to be inserted within the cavity formed by the frame and the two end plates. This modification enclosed the filtered sludge cake completely within the bag and, because the pressure drop is across the filter bag and the rubber seal gaskets are not under internal pressure, eliminated the necessity of double containment for this unit. The filtered cake is not open to the atmosphere during the unloading operation and, being neatly packaged within the filter bag (see Fig. 6.4), is in a convenient form from disposal in sealed containers. Pressure filters that employ this enclosed-bag innovation are not available commercially; therefore, additional design work would be required to develop the equipment for large-scale operation.

The average on-stream life of a filter bag was 10.17 hr, during which time an average of 23 lb of air-dried sludge cake, having a 50% moisture content, were collected in the bag. A drier cake is not required; excess water is removed merely to prevent dripping when the frame is rotated out of the end plates.
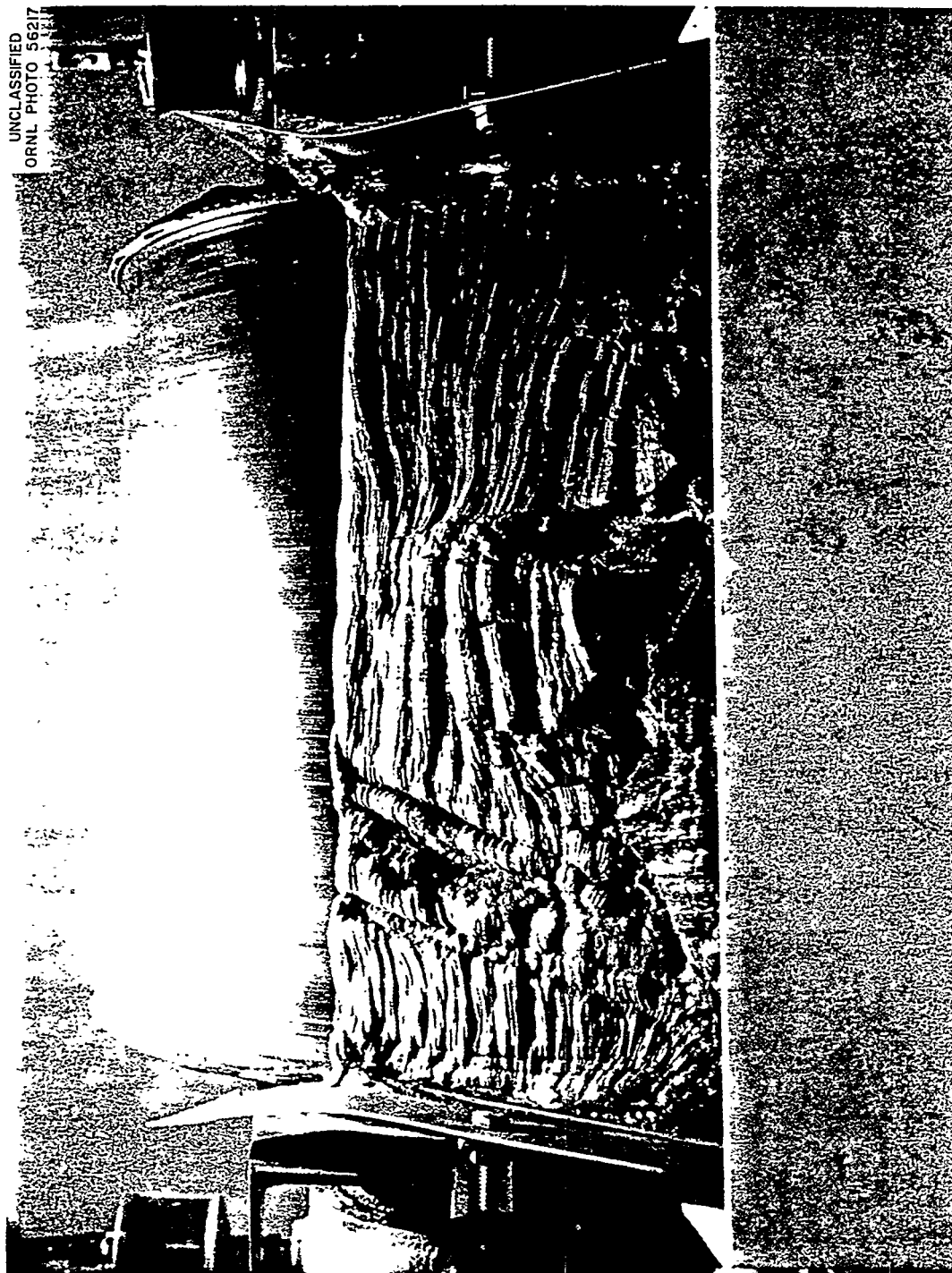
Fig. 6.3. Removal of sludge by vacuum filtration.

Fig. 6.4. Removal of sludge by pressure filtration.

6.4 Filtration of the Clarifier Effluent

Two filters in parallel were used to determine whether sand or anthracite was more effective for polishing the clarifier effluent prior to its entering the ion-exchange columns. Although sand reduced the filtrate hardness to 5 or 6 ppm, compared with 8 to 14 ppm for anthracite, sand is more difficult to backwash than the anthracite. Anthracite, which is lighter than sand, can be backwashed at lower flow rates. The filtrate was for all practical purposes, sparkling clear, having a turbidity of 1 to 2 ppm regardless of the filter medium. Based on the results, the anthracite method of filtration is recommended.

The 2-ft-diam Permutit vertical filters are of carbon steel, lined with Heresite, and each is equipped with a multiport valve to permit ease in filtering, backwashing, and rinsing. Each has a bed 33 in. deep and a cross-sectional area of 3.14 ft^2, which results in a flow rate of 3.2 gal min^{-1} ft^{-2} at the design feed rate of 10 gpm.

6.4.1 Sand Filtration

The sand filter is packed to a depth of 33 in. with 28- to 30-mesh quartz sand. After seven complete runs, or 28,000 gal of water, a hard, 1/8-in.-thick layer of calcium carbonate formed on top of the bed, which is shown in Fig. 6.5. This crust could not be broken with a slow 0.5 $\underline{M}$ HNO_3 backwash rate followed by backflushing with water at 7.7 gal min^{-1} ft^{-2}. A hole in the layer of calcium carbonate indicated that extensive channeling had occurred down through the bed. Further tests with new sand showed that an acid backwash should be made after every third water wash, thereby preventing crust formation. Accumulated solids increased the pressure drop across the filter to a maximum average of 10.4 psi before

Fig. 6.5. Formation of crust on sand filter.

hardness tests of the filtrate indicated the need for backwashing the sand bed. The average on-stream life of the sand filter was 72 hr before backwashing was necessary.

6.4.2 Anthracite Filtration

The anthracite filter had the same dimensions as the sand filter, but was packed to a depth of 33 in. with anthracite having an effective diameter size of 0.89 mm and a uniformity coefficient of 1.58.

Anthracite produced a filtrate that had a hardness of 8 to 14 ppm, which is only 5 ppm higher than that of the sand filtrate. In contrast to the sand, the anthracite bed shown in Fig. 6.6 had the same on-stream service as the sand. This bed was clean and uniform after backflushing with water at 7.7 gal min^{-1} ft^{-2}; however, after 12 runs there was evidence of a crust starting to form on the anthracite also, indicating that some acid backwashing is necessary but not as frequent as with the sand. Hardness tests indicate that the anthracite bed must be backwashed at a maximum average pressure drop of 7.3 psi.

The on-stream life of the anthracite is 80 to 98 hr, which is greater than the equivalent of 2,000 resin-bed volumes. Anthracite is satisfactory for polishing the clarifier effluent and, because it is more easily cleaned than the sand, the difference of only 5 ppm in the filtrates hardly warrants the use of sand, because the cost of each is about the same, $40 per ton.

6.5 Ion Exchange Step

The usefulness of the carboxylic-phenolic resin, Duolite CS-100, for removing the fission products remaining in the clarifier effluent was successfully demonstrated in two ion-exchange columns. Volumes of process
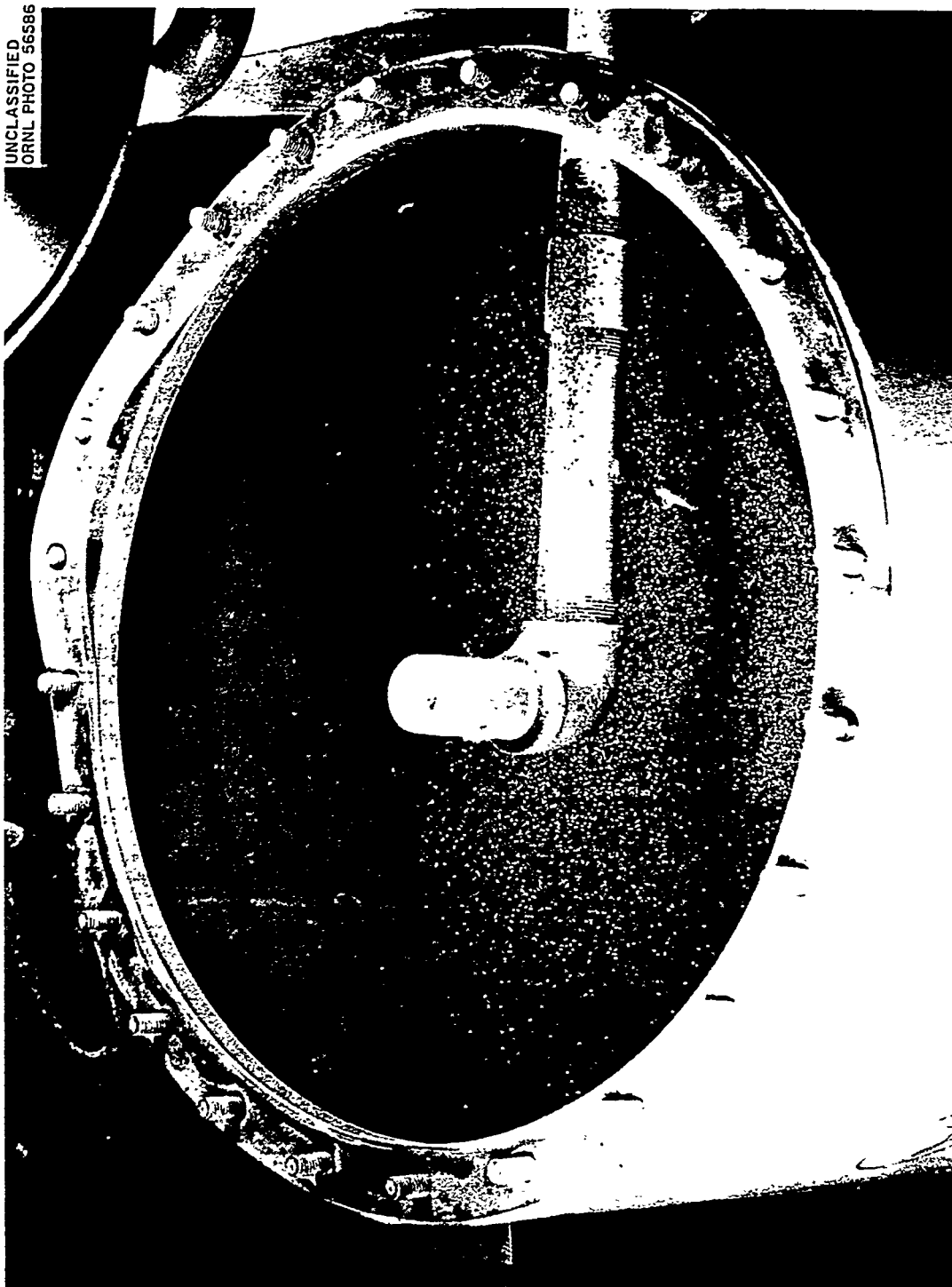
Fig. 6.6. Condition of antracite filter after seven runs.

wastes treated ranged from 1,520 to 3,130 bed volumes during the demonstration runs. In most cases, 1,500 to 2,000-bed-volume level was easily obtained with greater than 99.6% and 99.99% removal of cesium and strontium, respectively.

Each of the columns (Fig. 6.7) has a 3/32-in.-thick polyvinyl chloride lining over all wetted surfaces. Saran screening placed over all holes in the feed distributor and the bed-support plate retained the 28 gal of resin. A sight-glass pad placed at the midsection of the column permitted the observation of the resin bed. Polyvinyl chloride was selected because of the possibility of making demonstration runs with hydrochloric acid.

6.5.1 Sorption and Capacity of the Resin Beds

The maximum sorptive capacity of the resin beds for Sr^{90} and Cs^{137} was determined by extending the operating period beyond normal limits (2,000 bed volumes). In the case of HR-6, the cesium and strontium removal at the 2,000-bed-volume level agreed well with all process runs in the series. Cesium-137 was first to break through the resin bed after 2,300 bed volumes, with 50% breakthrough after 2,916 (Fig. 6.8). At this point, the plant effluent contained 11.5 dis min^{-1} ml^{-1} of Sr^{90}, representing 520% of the MPC_w. At 2,711 bed volumes, the total hardness (as calcium carbonate) of the effluent increased from less than 1 ppm to 3 ppm.

The sorption of the major isotopes, Cs^{137} and Sr^{90}, on the resin was a function of the hardness-removal efficiency of the precipitation-clarification equipment. The presence of phosphates in these low-activity wastes impedes the precipitation of calcium. Should the hardness not be effectively reduced in the head-end equipment, the calcium in the feed to the resin beds
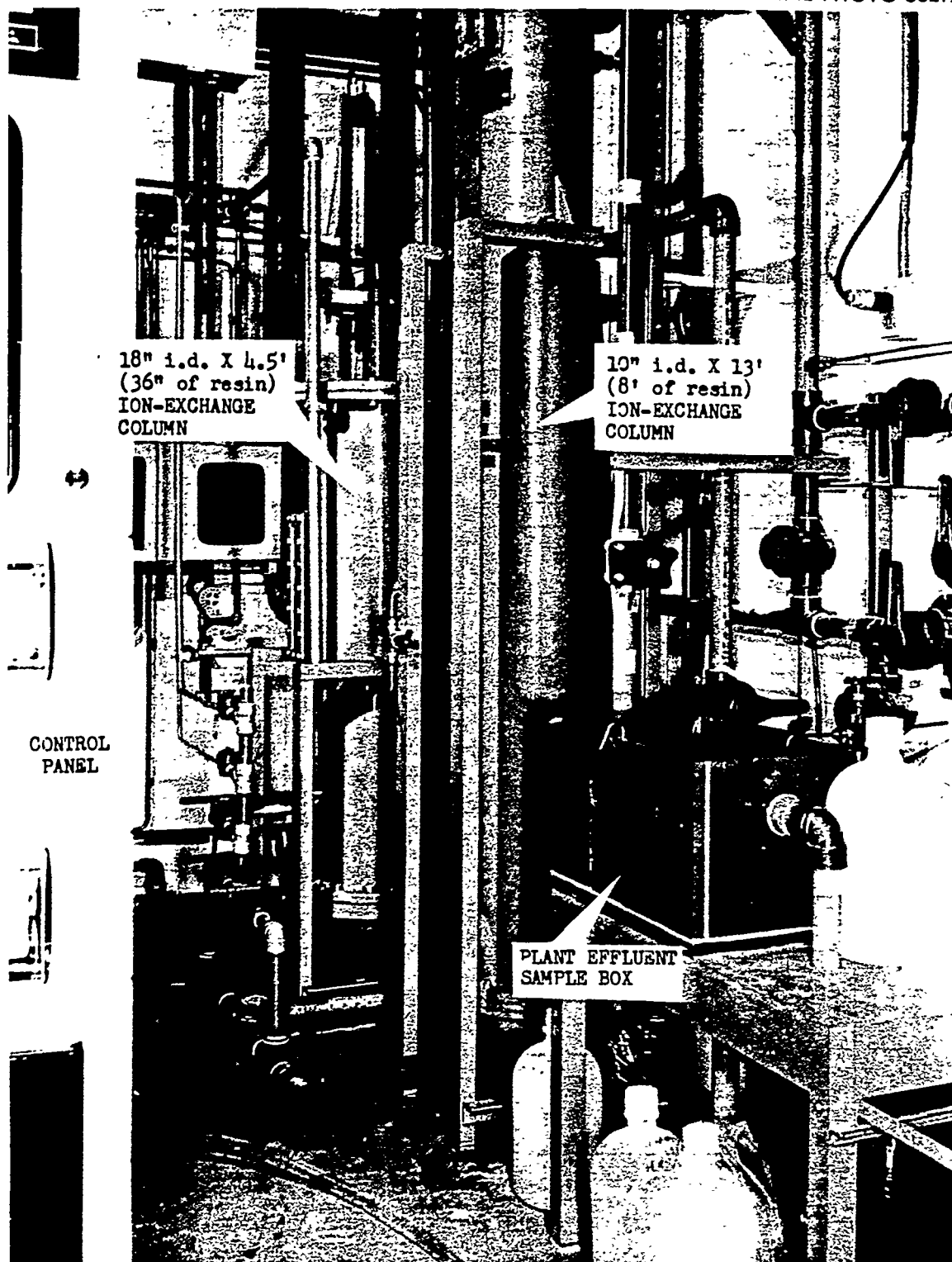
Fig. 6.7.   Low level waste treatment pilot plant — ion exchange columns.

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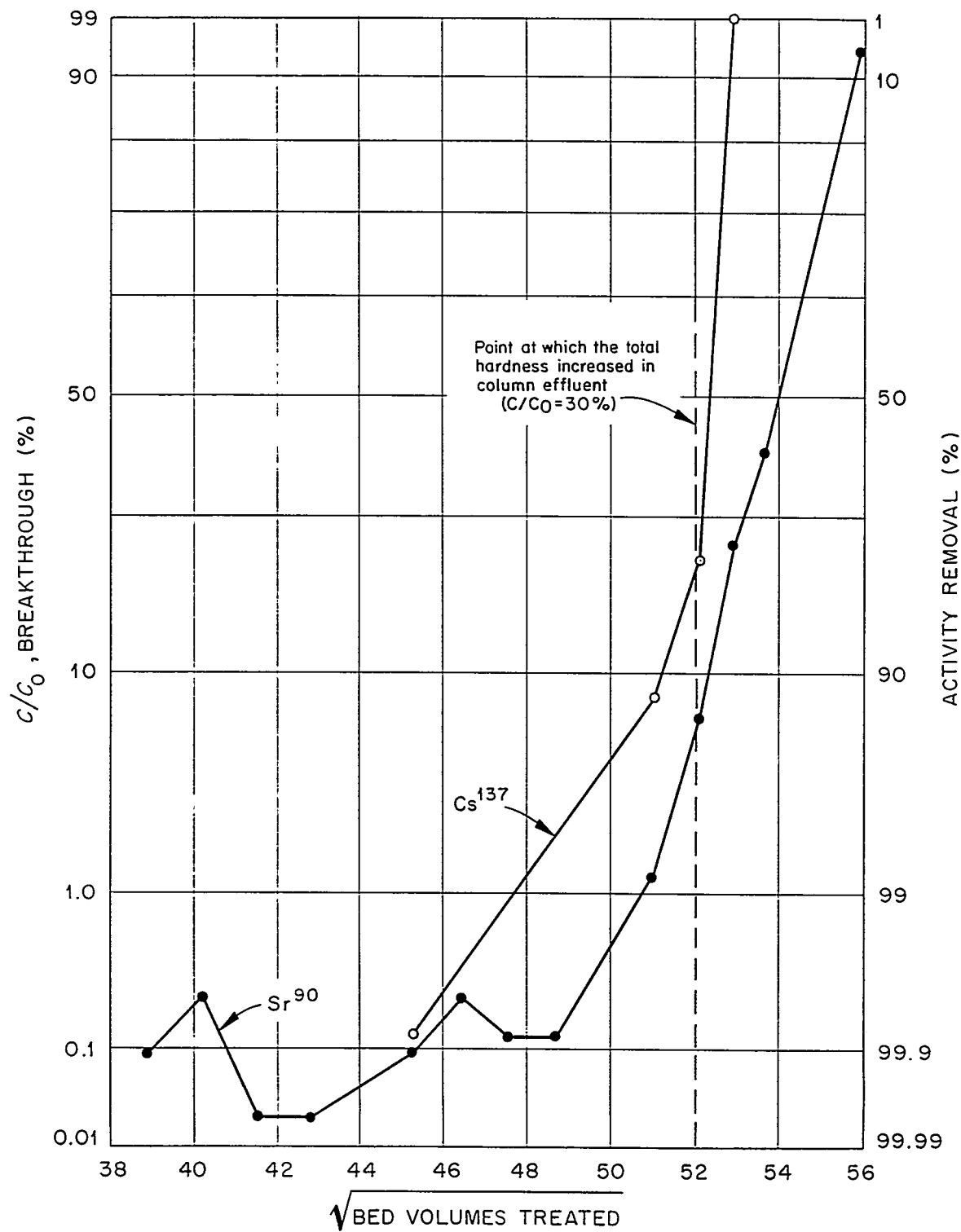
Fig. 6.8. Strontium-90 and cesium-137 breakthrough on Duolite CS-100 resin (run HR-6).

competes with the strontium and cesium for active resin sites. The use of 0.005 $\underline{M}$ Na_2CO_3 as an additive during periods when hexametaphosphate was present ensured the complete removal of the Sr^{90} by the resin but limited Cs^{137} removal (run HR-15, -16). The importance of hardness removal in the process initiated laboratory studies [5,7,8] to optimize the precipitation-clarification equipment and to study the overall effect of hexametaphosphate.

These studies showed that, with equipment modifications, a total hardness in the clarifier effluent of 1 to 3 ppm is obtainable in the presence of 0 to 2 ppm of hexametaphosphate, compared with the 8 to 14 ppm hardness in the filter effluent from the present equipment. As a result, it is predicted that more than 2,000 resin-bed volumes could be processed in the ion-exchange unit and still achieve decontamination factors as high as those obtained in the present study (see Sec 3.1). At hexametaphosphate concentrations of 2 to 4 ppm the addition of 0.005 $\underline{M}$ Na_2CO_3 will reduce the hardness to 1 to 3 ppm in the clarification step, but the number of bed volumes treated before cesium-breakthrough would be reduced by a factor of 2 because sodium and cesium compete for exchange sites. The strontium decontamination factor would not be affected.

Two ion exchange columns were tested to provide scale-up data for a full-scale plant. The 10-in.-diam by 8-ft-high bed was a scale-down (1/50 of full-plant-scale volume) at constant liquid velocity from the proposed plant beds. The 8-in.-diam by 32-in.-high bed had the same height-to-diameter ratio as the proposed plant and was designed to determine whether the data obtained from the 8-ft-high bed would require correction for a

lower ratio when scaling up to a full-scale bed. Three runs were made with the larger-diameter bed and the remainder with the smaller bed. No differences in process performance were noted in either case.

The same inventory of resin processed 87% of the solution handled during the program, with no noticeable decrease in sorption, elution, or regeneration characteristics.

6.5.2 Elution and Regeneration

The use of the split-elution procedure, which decreases the volume of spent solution leaving the process, was successfully demonstrated. In this procedure, five volumes of 0.5 $\underline{M}$ HNO_3 from the tail end of the previous elution is used as the initial eluant. The second five volumes, with a very low fission product concentration, is held in storage for a subsequent elution cycle. Typical elution curves (Fig. 6.9) graphically indicate that the bulk of the gross gamma, Sr^{90}, and Cs^{137} activities are removed in the first five volumes of eluant pumped through the bed.

The desorption of Cs^{137} and Sr^{90} was adequate at the end of each run. Of interest are the removal factors obtained by eluting the 10-in.-diam resin column after saturating the bed with cesium and strontium, as was the case during HR-6. Removal factors for Sr^{90} and Cs^{137} from the saturated resin bed were 3,140 and 1,750, respectively. This same bed was reused after regeneration, with no noticeable adverse effect on fission product removal or regeneration.

6.5.3 Gamma Scanning

The use of a movable gamma radiation probe through the length of the resin beds was effective for predicting resin-column breakthrough. Throughout each of the runs, the location of the maximum radiation band was noted
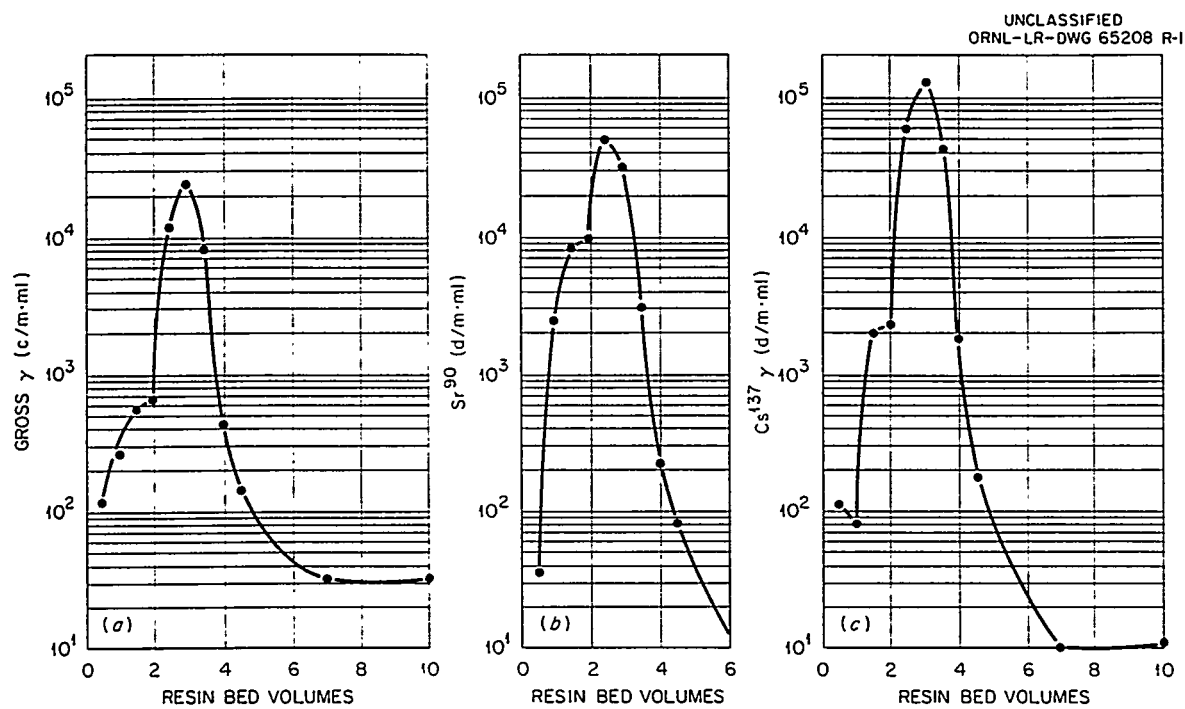
Fig. 6.9. Elution of (a) gross gamma, (b) Sr^{90}, (c) Cs^{137} from Duolite CS-100 resin with 0.5 $\underline{M}$ HNO$_3$ (HR-3).

as a function of the number of resin-bed volumes passed through the column.
Referring to a typical set of curves (Fig. 6.10), the formation of a 4-mr/hr
narrow band moved slowly to the 45- to 50-in. level. Shortly after 1,700
bed volumes, the hardness in the effluent from the polishing filter in-
creased suddenly, causing a slug of calcium ions to reach the resin bed.
After the flow was diverted to a spare filter and the hardness returned
to normal, a second activity band formed in the bed.

7. PROCESS COST ANALYSIS

Estimates of capital and operating costs for a 750,000 gal/day waste
treatment plant using the scavenging-precipitation ion-exchange process
have been reported[8] as $511,000 (including allowances for engineering
design and contingencies) and 54 cents per 1,000 gal of waste feed. These
estimates are based on a study that scaled up the data from laboratory
tests of the hydrochloric acid flowsheet.[6] The study indicated no signifi-
cant differences in costs between the hydrochloric and the nitric acid
flowsheets, but the nitric acid flowsheet appears more attractive for
ORNL application because it calls for reagents and materials of construc-
tion (stainless steels) that are more compatible with existing waste-
handling equipment and decontamination methods. Also, the nitric acid
flowsheet affords the possibility of eliminating the small regenerant-
acid waste evaporator required for the hydrochloric acid flowsheet by
utilizing the large stainless steel intermediate-activity waste evaporator
currently being designed for ORNL. The 54 cents per 1,000 gal does not
include amortization of the capital cost or the cost of storing the con-
centrated fission products from elution of the process ion exchange columns.
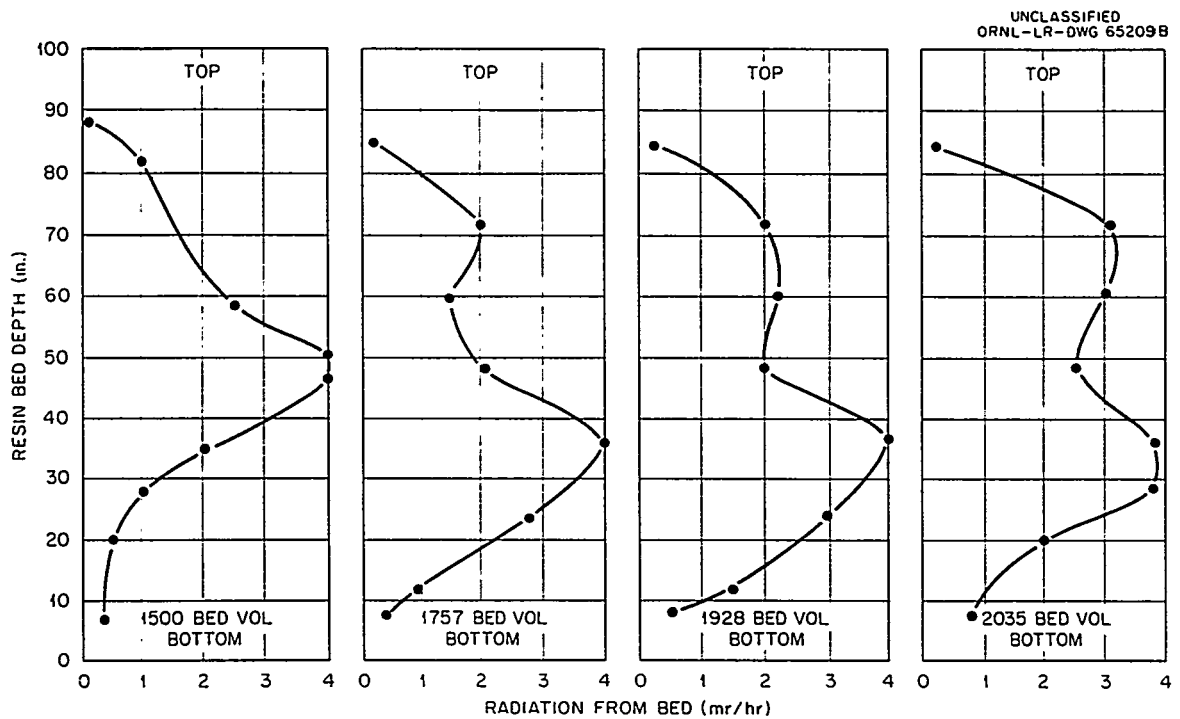
Fig. 6.10. Radiation profile of the resin during elution (run HR-3). Downflow elution; 10 in. dia column.

The process cost estimates have been revised to include additional information developed in the pilot-plant demonstrations of the nitric acid flowsheet. The pilot plant tests on ORNL low-activity waste confirmed the laboratory data, the major difference being that about 50% more sludge resulted from the scavenging-precipitation steps in the pilot plant. Figure 7.1 presents the 750,000 gal/day split-elution, nitric acid flowsheet on which the newer cost estimates are based. When the split-elution procedure is used, the ten volumes of 0.5 $\underline{N}$ HNO_3 required to elute the radioactive contaminants from the columns are split into two five-volume washes. This procedure results in a major saving by eliminating excessive waste handling, as most of the activity is removed in the first wash, allowing the second acid wash to be recycled.

The volume figures in Fig. 7.1 are those required for one cycle of ion-exchange column loading and regeneration. Since this flowsheet uses two ion-exchange columns, each employing two cycles per week, the weekly volumes can be found by multiplying each volume shown by four. This flowsheet is based on a throughput rate of 1,500 bed volumes (at 875 gal per bed volume) of low-activity ORNL waste per ion-exchange column cycle.

7.1 Capital Costs

Table 7.1 lists the capital equipment estimated for a full-scale scavenging-precipitation ion-exchange treatment plant for ORNL low-activity waste, the installed cost of which totals about $300,000; the customary allowances of 10% for engineering design and 20% for contingencies raise the cost to about $400,000. These figures were obtained from an engineering study in which vendors of the major equipment items were consulted about costs and applicability of the equipment proposed. The major
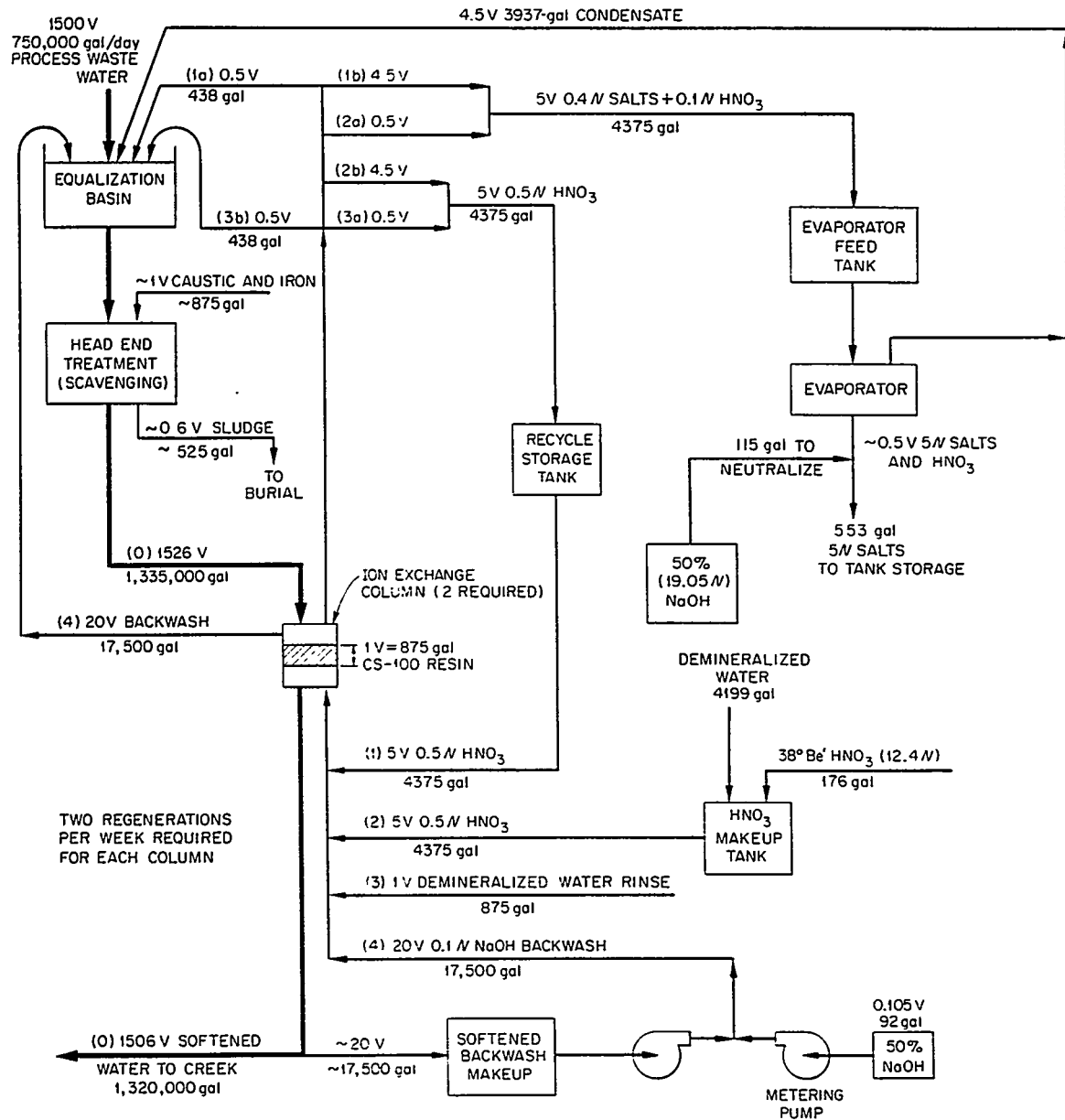
Fig. 7.1. Split-elution nitric acid flowsheet for the scavenging-precipitation ion-exchange process for treating low level waste.

Table 7.1 Estimated Capital Costs for 750,000-gal/day
Scavenging-Precipitation Ion-Exchange Low-Activity
Waste Treatment Plant (Split-Elution HNO_3 Flowsheet)

Item	Cost of Equipment	Cost of Installation	Total Cost
Sludge-blanket precipitator (29-ft diam, 15-ft height) and water meter	$25,000		
Automatic chemical feeders and blowdown	2,500	$9,200	
Concrete pad		1,200	$37,900
Flash mixer	500	200	700
Polishing filter, 250 ft^2 (U.S. Filter Company)	17,000	1,000	18,000
Eimcobelt vacuum sludge filter	7,000		
Vacuum receiver, wet vacuum pump, filtrate pump	1,200	2,500	10,700
4,000-gal carbon steel storage tank for 50% NaOH, 9 ft high, 9 ft in diam	3,300	700	4,000
2 ion exchange columns, 16 ft high, 4.5 ft in diam, Saran lined			36,000
234 ft^3 of Duolite CS-100 resin for columns at $22 per ft^3	5,150	500	5,650
Automatic controls and valves for ion-exchange columns	3,600 (installed)		3,600
10,000-gal recycle storage tank, 12 ft high, 12 ft in diam, Saran lined	11,000	1,500	12,500
10,000-gal evaporator feed tank, 12 ft high, 12 ft in diam, Saran lined	11,000	1,500	12,500
2,000-gal nitric acid storage tank, 7 ft high, 7 ft in diam, stainless steel	7,500	700	8,200
Continuous neutralizer	4,000	1,000	5,000
Piping			20,000
Instrumentation			5,000
Electrical Wiring			10,000
Building, 55 ft long, 44 ft wide, 24 ft high (55,000 ft3), at $2/ft^3			110,000
TOTAL			$299,750

difference in equipment from that used in the pilot-plant demonstrations is in the sludge and polishing filters. The U.S. Filter Co. tank-type pressure filter should serve as satisfactorily for the polishing filter as the sand and anthracite filters demonstrated in the pilot plant, but the difficulty with vacuum filtration experienced in the pilot plant might cause some concern about using the Eimcobelt vacuum filter listed in Table 7.1 for the sludge filter. Additional pilot-plant runs with a similar sludge filter are recommended to demonstrate this point. No evaporator for ion-exchange regenerant acid waste is included in this capital investment list, as use of the ORNL intermediate-activity waste evaporator, which has adequate capacity to handle this stream, is assumed.

7.2 Operating Costs

The estimated costs of operating a full-scale scavenging-precipitation ion-exchange treatment plant for ORNL low-level waste are presented in Table 7.2. Because these costs vary with ion-exchange column capacity, they are presented for column throughput rates of 2,000 down to 500 bed volumes (at 875 gal per bed volume) of low-activity waste fed to the process per column cycle. The pilot-plant runs routinely demonstrated throughputs of over 2,000 bed volumes of normal ORNL low-activity waste per cycle, achieving decontamination factors from Sr^{90} of over 1,000 and yielding effluents containing less than 3% MPC_w for Sr^{90} and Cs^{137}. The operating cost for this throughput rate is estimated to be 71.6 cents per 1,000 gal of waste treated. This cost includes chemicals, utilities, labor, maintenance, amortization, sludge burial, and evaporation, neutralization and storage of resin regenerant in steel tanks.

Table 7.2 Operating Costs for Various Throughput Rates of Ion-Exchange Columns, Scavenging-Precipitation Ion-Exchange Process for Low-Level-Waste Treatment

All costs are in cents/1,000 gal of waste fed to process

	Bed Vols			
	2,000	1,500	1,000	500
A. Fixed costs				
1. Utilities	1.0	1.0	1.0	1.0
2. Labor	16.4	16.4	16.4	16.4
3. Maintenance	7.0	7.0	7.0	7.0
4. Amortization	7.0	7.0	7.0	7.0
Total fixed costs	31.4	31.4	31.4	31.4
B. Chemical costs				
1. NaOH for pH adjustment	13.4	13.4	13.4	13.4
2. NaOH for column backwash	1.4	1.9	2.8	5.7
3. NaOH for regenerant waste neutralization	1.7	2.2	3.4	6.6
4. HNO$_3$ for resin regeneration	2.6	3.4	5.1	10.2
Total chemical costs	19.1	20.9	24.7	35.9
C. Waste-handling costs				
1. Evaporation of regenerant waste	8.8	11.7	17.6	35.1
2. Tank storage of regenerant waste concentrate	6.3	8.4	12.6	25.2
3. Burial of scavenging sludge	6.0	6.0	5.0	4.0
Total waste disposal costs	21.1	26.1	35.2	64.3
Total operating costs	71.6	78.4	91.3	131.6

The operating costs in Table 7.2 are broken down into fixed costs (utilities, labor, maintenance, and amorization) which do not vary with column throughput, chemical costs, and waste-handling costs. Labor cost is based on using one operator full time (168 hr/week, 52 weeks/year) at $3.00 per hour, with surcharges of 10% for supervision and 70% for overhead. Maintenance is estimated at 5% per year of the $400,000 capital investment, and amortization of this capital is spread over 20 years.

The chemical costs are based on prices of commodities delivered in bulk to ORNL according to existing contracts with chemical vendors. These prices agree well with those quoted in The Oil, Paint, and Drug Reporter. The only chemicals that appreciably affect costs are NaOH, HNO_3 and Na_2CO_3. All others including filter aids, replacement resin, and $FeSO_4$ amount to less than 0.1 cent per 1,000 gal of waste treated.

The cost of evaporating the regenerant acid from elution of the ion-exchange columns includes estimates of labor, steam, maintenance, and amortization for the ORNL intermediate-activity waste evaporator. Because of a recirculation system, condenser cooling water costs should be negligible. The total cost of evaporation is estimated at 3.5 cents/gal fed to the evaporator. The cost of storing the neutralized concentrate from this evaporation is estimated at 20 cents/gal stored in a new underground 1,000,000-gal steel tank that must eventually be built for an installation at ORNL after present tankage is filled. Neutralization costs are included in the NaOH cost, which assumes that all salts and acid in the regenerant solution require neutralization. The cost of sludge burial includes packaging the sludge in polyethylene-lined fiber drums, transportation to the ORNL burial ground, and 57 cents/ft^3 for burial ground space.

When ORNL low-activity waste contains greater than 2 ppm of hexameta-
phosphate (caused generally by heavy use of hexametaphosphate cleansers
in equipment decontaminations), the throughput rate of the ion exchange
column decreases to 500 bed volumes per cycle before strontium begins to
break through. The operating cost at this throughput rate is $1.32 per
1,000 gal (Table 7.2). Throughput can be increased to greater than 3,000
bed volumes before strontium breaks through (but only 900 bed volumes per
cycle before cesium breaks through) by the addition of 0.005 $\underline{M}$ Na_2CO_3 in
the head end (see Secs 2.3 and 6.5.1). This addition adds 10.6 cents per
1,000 gal to the operating cost, making a total of $1.02 per 1,000 gal for
a throughput rate of 1,000 bed volumes per cycle. These figures indicate
that it is cheaper to add carbonate to overcome phosphate-induced diffi-
culties than to reduce the number of bed volumes treated per ion-exchange
cycle. Of course, the most desirable solution to the problem is to elimi-
nate excessive use of hexametaphosphate cleansers.

The operating costs of the scavenging-precipitation ion-exchange
process compare quite favorably with the costs in other low-activity waste-
treatment processes of comparable effectiveness. The British quote $2.62
per 1,000 U.S. gal of waste treated in their least expensive process tried
at Harwell, which includes precipitation, adsorption on vermiculite, and
storage of sludge and spent vermiculite.[9] The British estimate includes
approximately the same items listed in Table 7.2. The lime-soda process
in use at ORNL has been estimated to cost 20 cents per 1,000 gal, exclusive
of maintenance, amortization, and sludge-burial costs.[10] Including these
would bring the cost to about 36 cents per 1,000 gal, about half of the
estimated cost for the scavenging-precipitation ion-exchange process.

However, the lime-soda process is so much less effective for strontium removal (decontamination factor of 7) compared to the scavenging-precipitation ion-exchange process (decontamination factor of over 1,000) that the two processes are not comparable. Use of the lime-soda process at ORNL imposes dependence on dilution of plant effluent in the Clinch River to achieve MPC_w values, and this process also is quite sensitive to the hexametaphosphate content of the waste.

8. CONCLUSIONS AND RECOMMENDATIONS

The results of the eight months' pilot-plant evaluation of the scavenging-precipitation ion-exchange process demonstrate that fission products can be removed adequately from large volumes of process wastes. Effluent streams leaving the process contained less than 3% of the MPC_w values for continuous occupational exposure for all isotopes. Decontamination factors for the major hazardous isotopes, Sr^{90} and Cs^{137}, ranged from 2,900 to 12,000 and 250 to 3,400, respectively, representing in most cases greater than 99.9% removal at the 1,500-to-2,000 resin-bed-volume level.

8.1 Conclusions

Based on the overall results of the pilot plant study the following conclusions may be made:

1. The scavenging-precipitation ion-exchange process is a workable method of decontaminating low-activity level wastes, using methods similar to standard water-treatment practices.

2. The economics of the flowsheet are such that wastes may be discharged without dilution at a cost of 72 to 78 cents per 1,000 gals (at the 1,500-to-2,000 bed-volume level) which is cheaper than other systems

capable of achieving the same degree of decontamination. A capital investment of $400,000 would be required to install a 750,000 gal/day plant at ORNL.

3. The process is superior to the lime-soda-clay system currently used at ORNL to treat low-activity-level wastes.

4. Although several equipment modifications would optimize the system, the equipment used was satisfactory.

5. The life and capacity of the resin, its regeneration characteristics, and scale-up data agreed with those predicted during early stages of the program. The demonstration of a split-elution cycle for reduction of waste volume and costs was significant.

8.2 Recommendations

The major recommendations are:

1. The use of hexametaphosphate should be administratively controlled because of the deleterious effect of this chemical on fission product removal. This effect is felt by both the lime-soda-clay and scavenging-precipitation ion-exchange methods of treating low-activity-level wastes. The use of 0.005 $\underline{M}$ Na_2CO_3 as an additive in the scavenging-precipitation ion-exchange process will permit satisfactory removal of Sr^{90} but will limit Cs^{137} removal for wastes containing up to 2 to 3 ppm PO_4^{3-}; however, additional costs would be incurred (11 cents/1,000 gal for the cost of the sodium carbonate at the 1,000 resin-bed-volume level).

2. Any improvement in the methods to improve hardness removal in the precipitation-clarification steps would increase the on-stream lives of the polishing filter and resin column. Several methods have been investigated in the laboratory and reported.[5,7] A redesign of the clarifier is recommended.

3. Anthracite filtration rather than sand filtration is recommended for polishing the clarifier effluent.

4. The applicability of the process to automation has been established. The use of radiation monitoring devices, and continuous hardness and turbidity monitors can do much to decrease the operating manpower requirements.

5. Additional study is recommended before a suitable method of sludge filtration is selected.

9. REFERENCES

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10. K. E. Cowser and R. J. Morton, "Radioactive Contaminant Removal from Waste Water: Evaluation of Performance," Journal of Sanitary Engineering Division, Proceedings of American Society of Civil Engineers (May 1959).

Table A1. Overall Removal of Activity from ORNL Waste

Run No.	Resin Bed Volumes	Gross β		Gross γ		Sr^{90}		Cs^{137}		Co^{60}		TRE β		$Zr\text{-}Nb^{95}$		Ru^{106}	
		DF	% Removed	DF	% Removed	DF	% Removed	DF	% Removed	DF	% Removed	DF	% Removed	DF	% Removed	DF	% Removed
HR-2	2000	30	96.70	44	97.7	2956	99.97	288	99.65	16	93.9	4	71.6				
HR-3	1500	46	97.80	25	96.1	6143	99.98	788	99.87								
HR-4	2086	42	97.60	10	89.9	2047	99.96	246	99.59	11	91.3	28	96.5				
HR-5	1959	37	97.3	16	93.8	4982	99.98	429	99.80	6	82.8	22	95.6				
HR-6[a]	1789					5588	99.96	2520	>99.96	4	74.3	31	96.8				
	1500					2316	99.96	544	99.80								
	2046					2316	99.96	12	91.50								
	2593					139	99.28	4	76.10								
	2711					29	96.54	6	82.30								
HR-7	3118	12	92.0	5	77.9	20	95.11	451	99.80	5	80.9	21	95.3				
HR-8[b]	2086	34	97.0	19	94.7	12160	99.99	153	99.35	121	91.9	24	95.8				
	300					8400	99.99	809	99.88								
	800					8400	99.99	688	99.85								
	1300					3360	99.97										
	1800					2800	99.96										
HR-9	2000	4	71.4	3	65.2	4200	99.98	3444	99.9	3	63.3	21	95.2				
	200					4098	99.97										
	800					8196	99.99										
HR-10	2131	26	95.4	14	93.1	8196	99.99	77	98.70	12	91.2	56	98.2				
HR-11[c]	1500	95	99.0	23	95.6	2880	99.97	200	99.50	18	94.4			151	99.3		
HR-12[d]	360	33	97.0	49	97.9	3150	99.97	116	99.1	7	86.3	649	99.8				
	860	28	96.4	39	97.4	3150	99.97	2	43.6	10	90.0	325	99.7				
	1500	3	60.3	2	52.3	15	93.46			5	81.4	3	64.6				
HR-13[c]	300					698	99.86	37.6	97.34	8	87.5					8.3	88.0
HR-14[d]	800					77	98.71		8.27	7	85.7	135	99.3	11	91.0	1.4	29.6
	1300					9	89.43		28.96	4	75.0	58	98.3			2.7	62.5
	1700					11	91.04			4	71.4	13	92.6			1.7	42.8
	1942					16	93.91			4	75.0	13	92.6	13	92.3	1.5	34.6
HR-15[e]	341					586	99.83	35.2	99.99			25	96.0				
	800					938	99.89	9.0	97.16								
< 1 ppm	1240	3	66.7	10	91.6	1563	99.94	13.3	88.85								
HR-16	1791	81	98.78	13	92.6	2344	99.96	76.0	92.50	6	84.0	442	99.78			2.7	63.33
	463					1032	99.90	8.9	98.68							5.0	80.00
	900					2580	99.96	1.2	88.82								
2 ppm	1350	20	95.0	4	71.1	2580	99.96		17.11	6	84.0	587	99.83			5.45	81.67
	1601					2580	99.96	1.6	39.47								

[a] Deliberate breakthrough run to ascertain Sr^{90}, Cs^{137}, and hardness limits.

[b] High-activity run. Co^{60} spill at ORNL, 1.7×10^3 dis min^{-1} ml^{-1} Co^{60} in feed.

[c] Precipitation-clarification equipment operated only.

[d] Excess (2 to 3 ppm) PO_4, in feed solutions, polishing filter breakthrough.

[e] Use of 0.005 $\underline{M}$ Na_2CO_3 as process additive.

Table A2. Radioactivity (Expressed as Percentage
of the MPC_w) in the Effluent from the
Scavenging--Ion Exchange Process

| Run No. | Bed Volumes | % of MPC_w[a] | | | | |
		Sr^{90}	Cs^{137}	Co^{60}	TRE β[b]	Ru^{106}
HR-2	2000	0.90	0.009	0.058	1.08	
HR-3	2086	0.90	0.18	0.112	0.12	
HR-4	1959	0.90	0.013	0.542	0.40	
HR-5	1789	0.90	0.011	0.333	0.20	
HR-6	3118	1.35	0.011	0.106	0.15	
HR-7	2086	0.45	0.029	0.067	0.07	
HR-8	2000	1.80	0	54.504	0.28	
HR-9	2131	< 0.45	0.067	0.216	0.07	
HR-10	1500	0.90	0.018	0.063	0.05	
HR-12	1500	463.96	8.820	0.270	1.55	
HR-14	1942	153.15	2.240	0.126	0.47	4.41
HR-15	179	2.70	0.830	0.144	0.07	0.45
HR-16	1600	0.90	2.070	0.072	0.02	0.99
Average		1.10[c]	0.30[c]	0.17[d]	0.35	1.95

[a]Based on the 168-hr MPC_w ("Maximum Permissible Body Burdens and Maximum Permissible Concentrations in Air and in Water for Occupational Exposure," NBS Handbook 69, June 5, 1959).

[b]Calculated as Y^{91}.

[c]Excludes runs HR-12, HR-14.

[d]Excludes runs HR-8.

Table A3. Fission Product Activities of Feed Solutions

Run No.	Gross β (c/m/ml)[a]	Gross γ (c/m/ml)	Sr^{90} β (d/m/ml)[b]	Sr^{89} (d/m/ml)	Co^{60} (d/m/ml)	Ru^{106} (d/m/ml)	TRE β[c] (c/m/ml)	Cs^{137} (d/m/ml)	Zr-Nb (c/m/ml)	I^{131} (d/m/ml)
HR-2	16.5	10.9	59.2		10.6	N.D.	5.6	11.5	< 0.2	< 1.0
HR-3	17.3	15.6	61.43	N.D.[d]	14.32	N.D.	5.10	39.42	3.42	N.D.
HR-4	21.00	19.77	99.79	3.74	34.99	N.D.	13.49	25.74	N.D.	N.D.
HR-5	30.60	330.07	111.76		14.39	N.D.	8.91	126.0	4.57	N.D.
HR-6	12.84	8.47	69.48		6.18		4.71	27.18	0.29	
HR-7	27.0	17.0	121.6		9.10		2.60	58.7	0.50	
HR-8	126.0	730.0	168.0		1650.0		9.0	275.0		
HR-9	18.06	14.4	81.96	4.96	29.5		5.63	23.04		
HR-10	28.50	9.0	57.6		12.4		10.60	16.0	1.0	
HR-12	33.5	19.5	157.50		16.10		6.49	69.50		
HR-13[e]	71.06[f]	7.89	76.48		7.52	1.93	9.69	10.43		
HR-14	50.00	17.70	46.2		5.2	15.0	18.0	14.5	7.8	
HR-15	18.0	29.0	46.89		6.87	1.70	13.25	49.32		
HR-16	36.2	9.0	51.6		5.0	12.0	17.6	15.2	4	

[a] Expressed as counts per minute per milliliter.

[b] Expressed as disintegrations per minute per milliliter.

[c] Counted at 10% geometry.

[d] Sought but not found.

[e] Precipitation-clarification equipment operated only.

[f] Counted at 40% geometry.

Table A4. Average Conditions of Process Streams

Run No.	Duration (hr)	Hardness as CaCO₃ (ppm)				Turbidity (ppm)			pH			
		Feed	Clarifier	Polishing Filter	Ion Exchange Column	Clarifier	Polishing Filter	Ion Exchange	Feed	Floc-culator	Clari-fier	Ion Exchange
HR-2	97	119.7	31.3	2.3	< 1.0	2.48	< 1.0	1.87	7.43	11.80	11.82	11.81
HR-3	96	123.6	57.3	8.41	< 1.0	4.96	0.75	2.13	6.93	11.86	11.89	11.85
HR-4	100	128.7	58.0	8.91	< 1.0	4.66	1.02	4.13	7.17	11.94	11.96	11.94
HR-5	71	138.8	46.0	9.50	< 1.0	3.99	1.25	3.61	7.85	11.96	11.97	11.96
HR-6	146	120.6	52.2	12.6	< 1.0	2.58	1.34	3.31	7.79	11.85	11.88	11.88
HR-7	94.5	122.1	64.1	12.5	< 1.0	4.06	1.76	2.47	7.77	11.89	11.91	11.90
HR-8	91	111.8	68.2	13.5	< 1.0	3.17	1.40	2.52	9.24	11.88	11.87	11.91
HR-9	98	108.65	67.0	5.2	< 1.0	5.53	2.02	3.04	8.23	11.84	11.84	11.86
HR-10	90	104.30	64.0	17.0	< 1.0	9.88	4.66	5.84	7.40	11.94	11.87	11.87
HR-11[a]	107	105.60	78.4			8.50			7.41	11.92	11.90	
HR-12	76	106.0	72.1	26.1	1.0	7.20	3.40	8.50	7.78	11.90	11.89	11.93
HR-13[b]	577	117.25	59.87	11.60	< 1.0							
HR-14	98	100.1	19.3	17.8	2.96	4.71	3.31	6.15	7.31	11.29	11.26	11.24
HR-15	98	96.3	13.3	10.7	< 1.0	11.99	8.76	8.86	7.60	11.38	11.36	11.34
HR-16	83.5	102.9 99.77	27.0 19.87	14.6 14.37	< 1.0 1.65	5.73	3.00	7.13	7.37	11.75	11.75	11.74
Average[c]	95.54	114.5	51.6	12.24	< 1.0	5.67	2.59	4.58	7.66	11.80	11.80	11.79

[a]Head-end treatment only (test run).

[b]Hardness test.

[c]Does not include run HR-11.

Table A5. Activity Removal Across Major Process Steps

Run No.	% Removal Across Precipitation Clarification-Filtration Steps						Across Ion Exchange Steps[a] (% Removed of Remaining from Previous Steps)						
	Gross β	Gross γ	Sr^{90}	Co^{60}	TRE	Cs^{137}	Gross β	Gross γ	Sr^{90}	Sr^{89}	Co^{60}	TRE	Cs^{137}
2	79.21	76.79	63.13	92.08	71.05		83.97	90.12	99.91		22.62	1.84	99.65
3	67.63	40.09	52.95	93.16	76.08	8.68	93.21	93.46	99.90		27.55	85.25	99.64
4	60.24	67.63	61.12	81.91	73.24	11.89	94.01	68.75	99.95	99.92	4.90	83.38	99.74
5	31.70	4.69	47.57	69.35	60.38	7.71	96.08	93.50	99.97		16.10	91.78	99.97
6	61.37	34.95	55.86	81.55	66.67	1.99	78.33	66.06	88.91		91.47	85.99	81.94
7	66.67	32.94	63.82	100.00	7.69	0.95	91.11	92.11	99.98			95.79	99.78
8	66.43	61.51	70.18	63.94	66.33	30.55	14.89	9.61	99.92		32.84	85.81	100.00
9	73.21	66.39	74.66	91.90	49.38	6.25	91.73	81.74	99.95		16.38	96.49	98.61
10	85.78	55.56	75.87	87.10	85.19	4.38	70.37	75.00	85.60		50.00	68.15	88.24
12	71.79	57.28	72.76	87.08	6.93	43.02	68.25	36.97	81.61		8.15	79.14	41.41
14	79.40	74.12	68.40	66.54	84.06	8.96	42.72	8.30	76.71		19.54	75.61	24.47
15	87.17	68.52	82.68	76.13	75.55	19.71	90.48	76.45	99.26		2.4	96.60	90.66
16	84.25	60.00	78.19	86.00	85.23	1.32	68.42	27.78	99.82		11.11	98.85	38.67

[a] Based on gamma spectrometry.

Table A6. Fission Product Activities of Plant Effluent

Run No.	Gross β (c/m/ml)[a]	Gross γ (c/m/ml)	Sr^{90} β[c] (d/m/ml)[b]	Sr^{89} (d/m/ml)	Co^{60} (d/m/ml)	Ru^{106} (d/m/ml)	TRE β (c/m/ml)	Cs^{137}[c] (d/m/ml)	Zr-Nb (c/m/ml)
HR-2	0.55	0.25	< 0.02	N.D.	0.65	0	1.60	0.04	0
HR-3	0.38	0.61	0.01-0.03	0	1.25	0	0.18	0.05-0.16	0
HR-4	0.50	2.00	0.02	0	6.02	0	0.60	0.06	0
HR-5	0.82	2.05	0.02	0	3.70	0	0.29	0.05	0
HR-6	1.05	1.87	0.03	0	1.18[d]	0	0.22	0.05	0
HR-7	0.80	0.90	0.01		0.75	0	0.11	0.13	0
HR-8[e]	36.00	254.00	0.04		605.00	0	0.43	0	0
HR-9	0.40	0.88	< 0.01	0	2.40	0	0.10	0.30	0
HR-10	0.30	0.40	0.02	0	0.70	0	0.07	0.08	0
HR-12[f]	13.30	9.30	10.30	0	3.00	Trace	2.30	39.2	0
HR-14	5.90	4.20	3.40		1.40	9.8	0.70	9.97	0.6
HR-15	0.22	2.15	0.06		1.60	0.99	0.11	3.70	
HR-16	1.80	2.60	0.02		0.80	2.20	0.03	9.20	

[a] Expressed as counts per minute per milliliter.

[b] Expressed as disintegrations per minute per milliliter.

[c] Isolated by radiochemical analysis; repurified and counted by low-activity analytical methods.

[d] After 300° bed volumes.

[e] High-activity run; Co^{60} spill at ORNL.

[f] PO_4 (3 ppm, 1500 bed volumes in feed solutions; premature breakthrough).

Table A7. Typical Format of FORTRAN Program

LOW LEVEL WASTE PILOT PLANT

RUN NO.HR-2

FEED   0.01 M NAOH   10 PPM FE

FEED RATE   10 GPM.   RUN DURATION   73 HOURS.   2000 VOLUME CHANGES.

CONTAMINANT	UNIT	FEED COMPOSITE	CLARIFIER OVERFLOW	RESIN FEED	PLANT EFFLUENT
GROSS BETA	C/M/ML	16.50	5.70	5.73	0.55
GROSS GAMMA	C/M/ML	10.90	2.25	2.25	0.25
SR-90 BETA	D/M/ML	59.12	25.15	21.60	0.02
SR-89	D/M/ML	-0.	-0.	-0.	-0.
CO-60	D/M/ML	10.60	0.68	0.84	0.65
RU-106	D/M/ML	-0.	-0.	-0.	-0.
TRE	D/M/ML	5.63	1.63	1.63	1.60
CS-137	D/M/ML	11.50	11.54	11.54	0.04
ZR-NB-95	D/M/ML	-0.	-0.	-0.	-0.
I-131	D/M/ML	-0.	-0.	-0.	-0.

RESULTS

CONTAMINANT	PRECIPITATION CLARIFICATION DECON. FACTOR	PERCENT REMOVED	ION EXCHANGE DECON. FACTOR	PERCENT REMOVED	OVERALL PROCESS DECON. FACTOR	PERCENT REMOVED
GROSS BETA	4.81	79.21	6.24	83.97	30.00	96.67
GROSS GAMMA	4.31	76.79	10.12	90.12	43.60	97.71
SR-90 BETA	2.71	63.13	1090.00	99.91	2956.00	99.97
SR-89	NONE OF THIS CONTAMINANT CONTAINED IN FEED.					
CO-60	12.62	92.08	1.29	22.62	16.31	93.87
RU-106	NONE OF THIS CONTAMINANT CONTAINED IN FEED.					
TRE	3.45	71.05	1.02	1.84	3.52	71.58
CS-137	1.00	-0.35	288.50	99.65	287.50	99.65
ZR-NB-95	NONE OF THIS CONTAMINANT CONTAINED IN FEED.					
I-131	NONE OF THIS CONTAMINANT CONTAINED IN FEED.					

Table A8.   FORTRAN List of the Computer Program for Data Handling

```
*TYPE(FORTRAN)
C      PILOTPLANTWASTE                                                   25500101
     1 DIMENSIONANAL(20),DIM(20),FEED(20),CLAR(20),RESIN(20),EFFL(20),    25500103
      1REST(20),DFPPT(20),REMPPT(20),DFION(20),REMION(20),                25500105
      2DFOA(20),REMOA(20)                                                 25500107
     2 I#1                                                                25500110
     3 READINPUTTAPE.2,100,NRUN,CAUST,.RON,NVOL,LONG,INRATE               25500112
     4 READINPUTTAPE10,101,ANAL(I),REST(I),DIM(I),FEED(I),CLAR(I),        25500114
      1RESIN(I),EFFL(I)                                                   25500116
     5 IF(FEED(I))8,6,6                                                   25500118
     6 I#I+1                                                              25500120
     7 GOTO4                                                              25500122
     8 I#I-1                                                              25500124
       NO#1
     9 WRITEOUTPUTTAPE9,102,NRUN,CAUST,IRON,NVOL,LONG,INRATE              25500201
    10 DO12K. ,I                                                          25500205
    11 WRITEOUTPUTTAPE9,103,ANAL(K),REST(K),DIM(K),FEED(K),               25500207
      1CLAR(K),RESIN(K),EFFL(K)                                           25500209
    12 CONTINUE                                                           25500211
    13 WRITEOUTPUTTAPE9,104                                               25500213
    14 DO29K#1,I                                                          25500301
    15 IF(FEED(K))28,23,16                                                25500303
    16 IF(RESIN(K))26,26,17                                               25500305
    17 DFPPT(K)#FEED(K)/RESIN(K)                                          25500307
    18 REMPPT(K)#(1.0-RESIN(K)/FEED(K))*100.0
    19 IF(EFFL(K))24,24,20                                                25500311
    20 DFION(K)#RESIN.K)/EFFL(K)                                          25500313
    21 REMION(K)#(1.0-EFFL(K)/RESIN(K))*100.0
    22 DFOA(K)#FEED(K)/EFFL(K)                                            25500317
    23 REMOA(K)#(1.0-EFFL(K)/FEED(K))*100.0
    31 WRITEOUTPUTTAPE9,108,ANAL(K),REST(K),DFPPT(K),
      1REMPPT(K),DFION(K),REMION(K),DFOA(K),REMOA(K)
    32 GOTO29
    24 WRITEOUTPUTTAPE9,105,ANAL(K),REST(K),DFPPT(K),REMPPT(K)            25500321
    25 GOTO29                                                             25500325
    26 WRITEOUTPUTTAPE9,106,ANAL(K),REST(K)                               25500401
    27 GOTO29                                                             25500403
    28 WRITEOUTPUTTAPE9,107,ANAL(K),REST(K)                               25500405
    29 CONTINUE                                                           25500407
    33 IF(NO-7)34,2,2
    34 NO#NO+1
    35 GOTO9
   100 FORMAT(2X,1A6,F8.0,4I8)                                           25500605
   101 FORMAT(3A6,4F8.0)                                                 25500607
   102 FORMAT(49H1              LOW LEVEL WASTE PILOT PLANT/35H0          25500411
      1                       RUN NO.1A6/24H0                  FEEDF7.25500413
      22,7H M NAOHI7,7H PPM FE/10H FEED RATEI5,20H GPM.   RUN DURATIONI5,25500415
      37H HOURS.I7,16H VOLUME CHANGES./58H0               FEED         25500419
      4 CLARIFIER   RESIN    PLANT/59H CONTAMINANT   UNIT   COMPOSITE  OV25500421
      5ERFLOW   FEED    EFFLUENT//)                                      25500425
   103 FORMAT(1H 2A6,1X,1A6,2F10.2,2F9.2)                                25500501
   104 FORMAT(1H0/39H0                           RESULTS/67H0            25500503
      1       PRECIPITATION           ION              OVERALL/67H       25500505
      2             CLARIFICATION        EXCHANGE          PROCESS/71 25500507
      3H              DECON.  PERCENT   DECON.  PERCENT   DECON.  P25500509
      4ERCENT/71H CONTAMINANT   FACTOR  REMOVED   FACTOR  REMOVED        25500511
      5FACTOR  REMOVED//)                                                25500515
   105 FORMAT(1H 2A6,2F9.2,39H   INFINITE  100.0     INFINITE  100.0)     25500517
   106 FORMAT(1H 2A6,57H  INFINITE  100.0       DOES NOT APPLY     INFINIT25500521
      1E  100.0)                                                         25500525
   107 FORMAT(1H 2A6,45H  NONE OF THIS CONTAMINANT CONTAINED IN FEED.)    25500601
   108 FORMAT(1H 2A6,2F9.2,2(F11.2,F9.2))
       END(0,1,0,0,0)                                                    25500609
```

Table A9.   Radiation Level

Run No.	Feed Line (mr/hr)		Flocculator (mr/hr)		Clarifier Bottom (counts/min x 10^3)		Clarifier Top (mr/hr)		Clarifier Side -Top (mr/hr)		Clarifier Side-M (mr/h)	
	Max.	Avg.	Max.	Avg.	Max.[a]	Avg.	Max.	Avg.	Max.	Avg.	Max.	A
HR-3	0.1	0.1	13.0	7.67	0.55	0.51	0.3	0.1^3	0.9	0.7	1.6	1
HR-4	0.2	0.1^6	3.0	1.8	5.5	3.9	0.2	0.1^5	0.5	0.2^4	1.8	0
HR-5	0.4	0.3^2	5.0	2.2	7.7	7.4	0.5	0.4^2	0.8	0.6^4	1.2	1
Overall	0.4		13.0		7.7		0.5		0.9		1.8	
Average		0.1^9		3.9		3.9		0.2^5		0.5		1
HR-8	4.0^a	3.25^a			6.0		20.0		6.5		20.0	

[a] Background.

r .	Clarifier Side -Bot. (mr/hr)		Surge Drum (mr/hr)		Polishing Filter (mr/hr)		Sludge Filter Open (mr/hr)		Sludge Filter Closed (mr/hr)		Resin Column (mr/hr)	
	Max.	Avg.	Max.	Avg.	Max.	Avg.	Max.	Avg.	Max.	Avg.	Max.	Avg.
	1.3	0.6	1.5	1.3			4.0	2.4	0.5	0.3	6.5	5.0
5	3.5	1.7[3]	5.5	3.5	2.0	1.2	9.0	4.8			0.2	0.1[4]
	5.0	5.0	6.0	4.4			12.0	10.0			1.2	0.4[7]
	5.0		6.0		2.0		12.0		0.5		6.5	
		2.4		3.1		1.2		5.7		0.3		1.9
	9.0		20.0								20.0	

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	Answering letter date

K-25

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K. W. Sommerfeld
J. J. Vogt
R. A. Winkel
File - NoRC

Subject Accidental Releases of
Hazardous Materials From
the Oak Ridge Gaseous
Diffusion Plant

As you requested, we have prepared an analysis of the effects of releases
of hazardous materials from the Oak Ridge Gaseous Diffusion Plant for
inclusion in your reply to the ERDA letter "Development of Safety Guidance
for ERDA Critical Facilities."

Please let us know if additional information is needed.

S. S. Stief

SSS:mj

Enclosure

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and 2-page attachment.

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The only hazardous materials (in terms of an off-site threat to public health) stored in significant quantities at the ORGDP are UF_6, anhydrous HF, and PCB's. The UF_6 is routinely stored as process inventory in the K-33, K-31, and K-29 process buildings, the K-1131 feed and tails withdrawal facility, the K-413 product withdrawal facility, and the K-1423 toll enrichment facility. The HF is stored in two 80,000-pound tanks located just inside the west perimeter fence and in two 5,000-pound tanks located adjacent to the fluorine plant in K-1131. The PCB's are contained in transformers and capacitors, the vast majority of which are located within the three process buildings.

Accidental releases of large quantities of these substances, such as might result from an earthquake or tornado, have been evaluated by ORGDP personnel. A brief summary of these evaluations, which assume worst-case conditions, is presented below.

ANHYDROUS HF RELEASE

The greatest potential for a significant HF release exists at the HF tank farm where as much as 160,000 pounds of anhydrous HF can be stored at any one time. In an effort to reduce the potential for such a release, modifications to this storage facility will be made as part of the "Containment of Oil and Hazardous Materials" project, which should commence in the near future. These modifications include the coating of the existing dike with an acid-proof epoxy to prevent an exothermic reaction of any released HF with the concrete, and the installation of a drain system that will discharge any liquid HF released into the dike into an underground storage tank, thus reducing the evaporative losses to the atmosphere. Since these changes have not yet been completed, two different releases have been hypothesized. In the first, the entire contents of both storage tanks would be instantaneously lost to the existing dike where it would remain until all of the HF had vaporized. In the second case, the proposed drain system would provide for the confinement of the released liquid in the underground tank.

Both cases assume the daytime temperature to be 90° F and the nighttime temperature to be 75° F. It was further assumed that the liquid HF in the storage tank was at a temperature of 90° F, so that about 8.75%, or 14,000 pounds, would flash to the atmosphere as soon as the tanks were ruptured. The major difference between the two cases is the time interval over which the release would occur, which subsequently translates into the total quantity of material released. In the first case, the entire contents (160,000 pounds) of both tanks would theoretically be vaporized in about two days. With the aid of the drainage and confinement system (Case 2), the 146,000 pounds of liquid HF contained in the dike would be transferred to the underground tank in about 10 minutes.

The consequences of both releases would be the same immediately following the accident, since both assume the instantaneous release of 14,000 pounds of HF to the atmosphere. According to dispersion calculations incorporating a five-mile-per-hour wind and stable atmospheric conditions, lethal concentrations of HF (greater than 100 mg/m^3) could exist at a distance of several miles downwind of the release. For the first case, where the contained liquid HF vaporized at an average rate of about 350 gm/sec, the lethal concentration could exist at a distance of about 0.5 mile from the release for a period of two days. In reality, the contained liquid could probably be transferred to a container relatively soon after the accident so that the two-day exposure period could be reduced. For the second case, the vaporization time would be limited to about 10 minutes, with the resulting ambient air concentration being essentially the same as for Case 1, except that they would exist for a much shorter period of time.

UF$_6$ RELEASE

Since the diffusion process system itself contains the largest quantity of UF$_6$, the most catastrophic release would be that resulting from breaching the system in several locations. This type of occurrence, while extremely remote, could conceivably result from a high intensity earthquake. However, since the size and thus the consequences of the release would be dependent upon the number of locations breached, it is virtually impossible to quantify either. It is apparent that one could postulate an extremely severe accident that could result in ambient air concentrations of HF far exceeding those hypothesized for the HF tank farm accident. The dispersion of the uranium is virtually unpredictable.

Of much less consequence, but somewhat more fixed with respect to quantities released, would be an accident involving a cylinder of UF$_6$. Such cylinders are routinely handled in and around the K-1131 tails withdrawal facility, the K-413 product withdrawal facility, and the K-1423 toll enrichment facility. Since the release of uranium from one of these cylinders would not present as great a problem as would the associated HF, the dispersion of the HF only was evaluated. For the extreme case involving the loss of the entire contents of a 14-ton cylinder of UF$_6$ in a period of about 15 minutes, lethal concentrations of HF could be evidenced as far as two miles downwind of the release during stable atmospheric conditions.

PCB AND OTHER CHEMICAL RELEASES

The ORGDP stores more than 140,000 gallons of PCB's in about 100 electrical transformers and over 13,000 capacitors. The largest quantity contained in any one location is 1,370 gallons, which is the capacity of one transformer. Since all but one of the transformers are located within buildings that will be secured to contain accidental releases from smaller systems, loss of the building would be necessary for a release to a nearby water stream. If this should occur, the PCB's which are heavier than and essentially insoluble in water, should settle to the bottom rather quickly. In view of the relatively small quantity released from one transformer, the consequences would not be expected to be catastrophic. The release from several transformers has not been evaluated but should not present an immediate health hazard.

Other chemicals stored at the ORGDP, such as HCl, H$_2$SO$_4$, Cl$_2$, ammonia, etc., should not pose a public health problem if released to the environment.

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